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An Analysis of Operator Splitting Techniques in the Stiff Case

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Operator splitting methods are commonly used in many applications. We focus here on the case where the evolution equations to be simulated are stiff. We will more particularly consider the case of two operators: a stiff one and a nonstiff one. This occurs in numerous application fields (e.g., combustion, air pollution, and reactive flows). The classical analysis of the splitting error may then fail, since the chosen splitting timestep Δt is in practice much larger than the fastest time scales: the asymptotic expansion $\Delta t \to 0$ is therefore no longer valid. We show here that singular perturbation theory provides an interesting framework for the study of splitting error. Some new results concerning the order of local errors are derived. The main result deals with the choice of the sequential order for the operators: the stiff operator must always be last in the splitting scheme.

Key Words: operator splitting; stiffness; singular perturbation; reduction of dynamical systems; reaction-diffusion PDEs; air pollution modelling.

1. INTRODUCTION

Operator splitting methods are widely used in many applications, such as Air Pollution Modeling [10, 17, 30], combustion [19], or general hyperbolic systems [3, 9, 15]. The first advantage of this approach is the use of specific tailor-made numerical solvers for each physical phenomenon to be integrated (e.g., advection, diffusion, and chemical production). The second advantage is the drastic reduction in CPU costs. Since the initial coupled system may be stiff (mainly due to chemical terms), the Method of Lines (MOL) could be highly expensive. The use of an implicit numerical scheme for the time integration is then recommended and leads to a large amount of algebraic manipulations since the dimension of the matrices to be inverted is typically given by the product of the number of variables, the chemical species, and the number of grid cells. Even if some structure can be exploited [5], the CPU cost is therefore rather large. As the only term inducing local coupling between

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species is the chemical production, an operator splitting method reduces the dimension of the matrices to be handled to the number of species.

The drawback of this method is however the error due to the uncoupling of operators. The classical analysis of such errors is based on asymptotic expansions of exponential operators in the linear case [29, 34] with respect to the splitting time step. The generalization to the nonlinear case [13, 22] proceeds in the same way with the help of Lie derivatives.

We will particularly focus on the case for which only one operator induces stiffness (which is often the case in practice). We then want to integrate a linear evolution system under a singular perturbation form

$$\frac{dz}{dt} = \left(\frac{\chi}{\varepsilon} + T\right) z, \qquad z(0) = z_0, \tag{1}$$

where ε is a small positive parameter tending to 0. ε measures the stiffness of the operator $\frac{\chi}{\varepsilon}$ and can be defined as the ratio of the fast time scales to the slow time scales.

First-order splitting schemes are easily defined by first integrating one operator and then the other on time intervals of length Δt (the so-called splitting time step). The usual analysis in the linear case leads to a local splitting error *le*, which is a second-order error in Δt

$$le \sim \left(\frac{\chi T - T\chi}{\varepsilon} z_0\right) \Delta t^2. \tag{2}$$

Such an analysis is particularly hazardous in the stiff case, unless the splitting time step actually tends to zero in order to meet $\Delta t \ll \varepsilon$. As modelers are supposed to use efficient solvers for the time integration (that is to say implicit schemes) this cannot be satisfied in practice since this would be equivalent to stability requirements similar to those needed for explicit schemes. Therefore we have in practice

$$\Delta t \gg \varepsilon. \tag{3}$$

The usual analysis leading to Eq. (2) should indicate that the error grows with stiffness. Splitting errors are however rather low in practical situations (the errors due to the treatment of boundary conditions being from now on neglected). The main reason for that is actually the stabilizing effect of the stiffness: the theoretical counterpart of numerical stiffness is indeed the existence of an underlying reduced model [14, 21, 24, 28]. Another point of view is to say that z_0 lies in certain subspaces (even in the split model), such that the terms of magnitude ε^{-1} disappear in the previous formula.

We will therefore follow an alternative analysis:

- we first compute up to first order in ε the reduced models for the coupled and the split schemes,
- we then compare with Δt tending to 0.

The main advantage of this double-limit approach is that the perturbation terms in $(\Delta t)/\varepsilon$ can be avoided.

Let us mention some related works to conclude. Such coarse integration has already been studied for hyperbolic systems with a stiff relaxation term [2, 3, 9, 15]. The purpose was however slightly different and focused mainly on grid refinement and shock tracking. A similar analysis has been proposed in [1, 12] but was restricted to the oscillatory case, which

is not pertinent for applications including phenomena such as chemical kinetics. Let us notice moreover that the focus was not put on the crucial role of the sequence of integration. The present article deals with the general case, both oscillatory and nonoscillatory, in contrast to [1, 12], although the emphasis is put on the nonoscillatory case.

After having defined some splitting techniques (Section 2), we investigate the proposed approach in the linear case (Section 3). Some numerical tests are then presented in the linear case (Section 4) and for Reaction-Diffusion Partial Differential Equations issued from Air Pollution Modeling (Section 5).

2. SPLITTING TECHNIQUES

Let us consider the following linear evolution system:

$$\frac{dz}{dt} = Az + Bz, \qquad z(0) = z_0, \qquad z \in \mathbf{R}^n, \tag{4}$$

where A and B are linear operators.

Let us first define the classical splitting schemes on $[0, \Delta t]$, with Δt the splitting time step.

2.1. First-Order Schemes

• *(A–B) Splitting*

We consider the scheme

$$\begin{cases} \frac{dz^*}{dt} = Az^*, \quad z^*(0) = z_0 \quad \text{on } [0, \Delta t] \\ \frac{dz^{**}}{dt} = Bz^{**}, \quad z^{**}(0) = z^*(\Delta t) \quad \text{on } [0, \Delta t], \end{cases} \tag{5}$$

where the final value is given by $z^{**}(\Delta t)$.

• *(B–A) Splitting*

This method is defined by changing the sequence of successive integration for A and B.

Such schemes are first-order schemes with respect to the splitting time step Δt. For instance the local error for the (A–B) splitting is given by

$$le = (exp(B\Delta t)exp(A\Delta t) - exp((A + B)\Delta t))z_0. \tag{6}$$

The usual study of this error is performed by asymptotic expansion and leads straightforwardly to

$$le = \frac{BA - AB}{2}\Delta t^2 z_0 + O(\Delta t^3). \tag{7}$$

The global error is then a first-order error with respect to Δt unless A and B commute.

2.2. Second-Order Schemes

In order to improve the accuracy, Strang [29] proposed to symmetrize the splitting scheme. A first approach is to take

$$z(\Delta t) = \frac{1}{2}(z_{AB}(\Delta t) + z_{BA}(\Delta t)), \tag{8}$$

where z_{AB} and z_{BA} are the solutions computed, respectively, with the schemes (A–B) and (B–A). This is a rather expensive technique since each operator has to be integrated twice on the splitting interval.

Another less time-consuming approach is then defined by the scheme

$$\begin{cases} \frac{dz^*}{dt} = Az^*, \quad z^*(0) = z_0 \quad \text{on } \left[0, \frac{\Delta t}{2}\right] \\ \frac{dz^{**}}{dt} = Bz^{**}, \quad z^{**}(0) = z^*\left(\frac{\Delta t}{2}\right) \quad \text{on } [0, \Delta t] \\ \frac{dz^{***}}{dt} = Az^{***}, \quad z^{***}(0) = z^{**}(\Delta t) \quad \text{on } \left[0, \frac{\Delta t}{2}\right]. \end{cases} \tag{9}$$

The final value is then given by $z^{***}(\frac{\Delta t}{2})$. We will name (A–B–A) this scheme and a scheme (B–A–B) can be derived in the same way. The interesting point is that the local error associated with this scheme is then

$$le = \left(exp\left(A\frac{\Delta t}{2}\right) exp(B\Delta t) exp\left(A\frac{\Delta t}{2}\right) - exp((A+B)\Delta t) \right) z_0 \tag{10}$$

and an asymptotic expansion leads easily to

$$le = O(\Delta t^3). \tag{11}$$

This scheme is then a second-order scheme and is used in practice for many applications.

Remark [Higher order terms]. Higher order terms can be computed with the use of the Baker–Campbell–Hausdorff formula. We refer for instance to [8, 13]. Numerous extrapolation techniques have been proposed for improving the accuracy of such schemes [32].

Remark [Extension to the nonlinear case]. Such an analysis can be performed in the nonlinear case with the use of the Lie operator formalism [13, 22]. We do not focus here on this point since we instead want to stress the influence of the stiffness on such analysis. The linearity will not play any role in the following analysis but it will clarify the computations. Nonlinear operators (associated with chemical kinetics) will however be taken into account in the numerical examples (Section 5).

2.3. No Time Splitting and Source Splitting

Another less common splitting scheme has been already proposed, mainly for Air Pollution Modeling [11, 30]. It is a slight modification of a first-order scheme as one operator (let us say A) is supposed to be nonstiff. In order to avoid transient phases due to stiffness, the initial conditions for the second substep are not modified but a source term must be added in order to take into account the first substep.

The proposed scheme can be put in the form

$$\begin{cases} \frac{dz^*}{dt} = Az^*, \quad z^*(0) = z_0 \quad \text{on } [0, \Delta t] \\ \frac{dz^{**}}{dt} = Bz^{**} + \frac{z^*(\Delta t) - z_0}{\Delta t}, \quad z^{**}(0) = z_0 \quad \text{on } [0, \Delta t]. \end{cases} \tag{12}$$

The final value is then $z^{**}(\Delta t)$. Let us notice that this is equivalent to an explicit integration of the nonstiff operator A.

We will call this scheme (*NTS*) (in the terminology of [30] where such a scheme is referred as "No Time Splitting").

Let us compute the local error for this scheme. We have straightforwardly

$$z_{NTS}(\Delta t) = exp(B\Delta t)\left[I + \int_0^{\Delta t} exp(-tB)\, dt\, \frac{exp(A\Delta t) - I}{\Delta t}\right] z_0. \tag{13}$$

We use the asymptotic expansion of the exponential operator and we obtain directly

$$z_{NTS}(\Delta t) = \left[I + (A + B)\Delta t + \frac{A^2 + B^2 + BA}{2}\Delta t^2 + O(\Delta t^3)\right] z_0 \tag{14}$$

and

$$le = z_{NTS}(\Delta t) - exp((A + B)\Delta t)z_0 = -\frac{AB}{2}\Delta t^2 z_0 + O(\Delta t^3), \tag{15}$$

which confirms the first-order nature of this scheme.

Remark [Why we do not consider the other (*NTS*) scheme]. Another (*NTS*) scheme can of course be defined by reversing the sequence. We will not consider this scheme. The first immediate reason is that it is well understood that integrating explicitely a stiff operator (let us say B) makes no sense. The importance of finishing the integration with the stiff operator B will be stressed in the next section.

3. AN ALTERNATIVE APPROACH FOR SPLITTING ERRORS

3.1. Why the Classical Error Analysis May Fail in the Stiff Case

Let us now focus on the case

$$A = \frac{\chi(\varepsilon)}{\varepsilon}, \qquad B = T, \tag{16}$$

where $\chi(\varepsilon) = \chi_0 + \varepsilon\chi_1$ is a slow–fast operator which induces the stiffness (for instance chemical kinetics) and T is a slow operator (for instance the spatial discretization of diffusion).

χ_0 and χ_1 denote, respectively, the fast and slow parts of χ. ε is as expected a small positive parameter supposed to tend to 0.

The key point is that

$$\Delta t \gg \varepsilon \tag{17}$$

is met in practice since tailored algorithms are used for the time integration of the stiff operator A. Let us mention that $\Delta t \sim \varepsilon$ or $\Delta t \ll \varepsilon$ are exactly the stability requirements for explicit schemes which are actually avoided in order to improve the CPU performance. We will name this practical case the *coarse case*.

Some immediate remarks can then be mentioned on the basis of the usual local error for first-order schemes (e.g., $(T - \chi)$) which yields

$$le \sim \left(\frac{\chi}{\varepsilon} T - T \frac{\chi}{\varepsilon} \right) \frac{z_0 \Delta t^2}{2} \tag{18}$$

by using (7) although higher order terms may be of course much larger.

1. The sequential order of integration should have no influence on accuracy in the usual analysis: the schemes $(T - \chi)$ and $(\chi - T)$ should then have the same behavior. This is rather surprising in the stiff case for which one operator is associated with slow dynamics (T) and the other one with slow and fast dynamics (χ). If χ is only concerned with fast dynamics $(\chi_1 = 0)$ one could advocate to first integrate the fast dynamics (χ) and then the slow ones (T).
2. Such a local error would indicate that the error is an increasing function in the stiffness ratio ε (with a fixed Δt). This is in total contradiction with the widespread argument that splitting schemes have to be used for well separated timescales (related to $\varepsilon \to 0$).

By anticipating the first example in Section 5, Fig. 1 gives the relative error for the splitting schemes $(\chi - T)$ and $(T - \chi)$ as a function of the splitting time step. χ describes here a particular kinetic scheme arising in atmospheric chemistry and T describes the discretization of a monodimensional diffusion term.

As a result of this test the sequence seems to be important. The scheme we naively advocated in our first remark is therefore not the good one: one has rather to begin the splitting process with the slow dynamics (T) and to end with the fast dynamics (χ)!

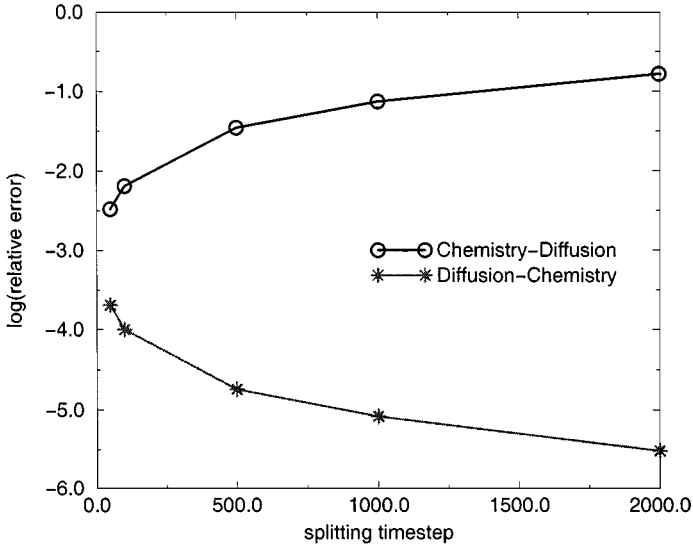

FIG. 1. Some surprising results (Example 5.1).

Our second remark applies to the "good splitting" $(T-\chi)$ after having noticed that $\Delta t \to \infty$ can be "replaced" with $\varepsilon \to 0$ in a scaling analysis. This is a rather surprising behavior: the bigger the splitting time step is, the more accurate the "good splitting" $(T-\chi)$ is!

3.2. Reduction

In order to investigate such behavior we want now to replace the usual analysis by an alternative analysis. As the main difficulty is provided by stiffness, we compute the underlying "reduced" model [14, 21, 23, 24, 28], which approximates the exact solution up to first-order in ε. We will then compare the reduced models respectively associated with the splitting schemes and with the exact solution which for both operators are integrated simultaneously.

Another point of view is that we want to assess the asymptotic behavior of the splitting schemes with respect to an increasing stiffness (as ε tends to 0): do the splitting schemes preserve the same behavior as the exact solution?

We refer to [14, 21, 23, 24, 28] for the theoretical background and more details on reduction. We will only mention the kernel of this theory.

Let us recall that the stiff operator χ is partitioned as

$$\chi = \chi_0 + \varepsilon \chi_1, \tag{19}$$

where χ_0 and χ_1 stand, respectively, for the fast and slow parts.

Assumption: slow–fast structure for χ_0. We assume the "semi-stability" of the fast operator χ_0:

- the eigenvalues of χ_0 are either null or have a strictly negative real part,
- the following subspace decomposition holds:

$$\mathbf{R}^n = ker(\chi_0) \oplus R(\chi_0). \tag{20}$$

Let us stress the fact that such an (apparently) technical assumption is usually met for chemical kinetics [14, 24].

Let $n-p$ be the dimension of $ker(\chi_0)$. Such an hypothesis justifies the existence of a change of basis

$$M = \begin{bmatrix} P \\ Q \end{bmatrix}, \qquad M^{-1} = [\bar{P} \quad \bar{Q}], \tag{21}$$

where M, P, and Q are, respectively, $n \times n$, $(n-p) \times n$, and $p \times n$ matrices. $\bar{P}$ and $\bar{Q}$ are, respectively, $n \times (n-p)$ and $n \times p$ matrices. P is chosen such that $P\chi_0 = 0$ that is to say that the lines of P are given by a basis of $R(\chi_0)^{\perp}$. A consequence is that the columns of $\bar{Q}$ define a basis of $R(\chi_0)$ since $P\bar{Q} = 0$.

Let us now write

$$x = Pz, \quad y = Qz, \qquad x \in \mathbf{R}^{n-p}, \qquad y \in \mathbf{R}^p. \tag{22}$$

This change of basis is a decoupling transformation: x is a slow variable while y is a fast variable since:

$$M \frac{\chi_0}{\varepsilon} M^{-1} = \begin{bmatrix} 0 & 0 \\ \frac{C}{\varepsilon} & \frac{-D}{\varepsilon} \end{bmatrix}, \tag{23}$$

where $-D = Q\chi_0\bar{Q}$ is a strictly stable square matrix (whose dimension is p). This point is easy to prove: let $Q\chi_0\bar{Q}u = 0$ with $u \in \mathbf{R^n}$. We then have $\bar{Q}Q\chi_0\bar{Q}u = 0$. By using $\bar{P}P + \bar{Q}Q = I$ and $P\chi_0 = 0$, this implies $\chi_0\bar{Q}u = 0$. That is to say that $\bar{Q}u \in ker(\chi_0)$. As $\bar{Q}u \in R(\chi_0)$ we have $u = 0$ and this ends the proof.

In the following we will refer to this new basis as the *partitioned basis*. It is now easy to derive the evolution in this basis:

$$\begin{cases} \frac{dx}{dt} = P\chi_1(\bar{P}x + \bar{Q}y) + PT(\bar{P}x + \bar{Q}y) \\ \varepsilon\frac{dy}{dt} = Cx - Dy + \varepsilon Q\chi_1(\bar{P}x + \bar{Q}y) + \varepsilon QT(\bar{P}x + \bar{Q}y) \end{cases} \tag{24}$$

with the prescribed initial conditions

$$x(0) = Pz(0), \qquad y(0) = Qz(0). \tag{25}$$

In our analysis we will first assume that ε tends to zero with a fixed Δt (coarse case for time splitting). The direct application of classical results of singular perturbation theory [31] ensures the following result we give in a formal way.

THEOREM 3.1. *Up to a transient phase* (*of length* $O(\varepsilon)$), *the initial system* (24) *can be approximated up to first order in* ε *by the differential-algebraic system* (*of index* 1):

$$\frac{dx}{dt} = P\chi_1(\bar{P}x + \bar{Q}y) + PT(\bar{P}x + \bar{Q}y), \qquad 0 = Cx - Dy \tag{26}$$

with the modified initial conditions (*consistent with the algebraic constraint*)

$$x(0) = Pz(0), \qquad y(0) = D^{-1}Cx(0). \tag{27}$$

For $t \in [0, \Delta t]$ *the error associated with the reduced solution* (26) *can be put in the form*

$$O(\varepsilon) + O\left(exp\left(-\frac{\gamma\Delta t}{\varepsilon}\right)\right), \tag{28}$$

where $\gamma > 0$ *depends only on* D *and indicates the rate of convergence toward the reduced model.*

Such an approximation is of course valid only in the coarse case ($\Delta t \gg \varepsilon$).

From now on the approximation symbol $\sim$ will hold for the error associated with reduction and its magnitude is given by (28).

If we go back to the initial basis, the reduced solution associated with the exact model (that is to say without splitting) is then easily given by

$$z_{WS}(\Delta t) \sim (\bar{P} + \bar{Q}D^{-1}C)exp[P(\chi_1 + T)(\bar{P} + \bar{Q}D^{-1}C)\Delta t]Pz_0, \tag{29}$$

since $z = \bar{P}x + \bar{Q}y$. From now on the subscripts "WS" (Without Splitting) will stand for the case for which the operators are integrated simultaneously..

We will write for convenience in the sequel

$$K = \bar{P} + \bar{Q}D^{-1}C, \qquad S = P\chi_1 K \tag{30}$$

K is a rectangular matrix defined by the change of variables from the reduced model to the initial basis. S is the slow term induced by the stiff operator in the reduced model. With these notations we straightforwardly have the following lemma:

LEMMA 3.1 (Reduced model for the exact solution WS).

$$z_{WS}(\Delta t) \sim K\, exp[(S + PTK)\Delta t]Pz_0 \tag{31}$$

3.3. Computing a Reduced Solution for the Splitting Schemes

Let us now calculate the reduced solution for each of the splitting schemes.

LEMMA 3.2 (Reduced model for different splitting schemes).

$$\begin{cases} z_{T-\chi}(\Delta t) \sim K\, exp(S\Delta t)P\, exp(T\Delta t)z_0 \\ z_{\chi-T}(\Delta t) \sim exp(T\Delta t)K\, exp(S\Delta t)Pz_0 \\ z_{\chi-T-\chi}(\Delta t) \sim K\, exp\left(S\frac{\Delta t}{2}\right)\, P\, exp(T\Delta t)K\, exp\left(S\frac{\Delta t}{2}\right)Pz_0 \\ z_{T-\chi-T}(\Delta t) \sim exp\left(T\frac{\Delta t}{2}\right)\, K\, exp(S\Delta t)P\, exp\left(T\frac{\Delta t}{2}\right)z_0 \\ z_{NTS}(\Delta t) \sim K\, exp(S\Delta t)\left[P + \int_0^{\Delta t} exp(-tS)\, dt\, P\, \frac{exp(T\Delta t)-I}{\Delta t}\right]z_0 \end{cases} \tag{32}$$

Proof. We keep the same notations as in Section 2 for defining the substeps of the splitting methods. We will omit some computations left to courageous readers (if any).

For the $(T-\chi)$ splitting we have for the second step (by using (24) with $T=0$):

$$\begin{cases} \frac{dx^{**}}{dt} = P\chi_1(\bar{P}x^{**} + \bar{Q}y^{**}) \\ \varepsilon\frac{dy^{**}}{dt} = Cx^{**} - Dy^{**} + \varepsilon Q\chi_1(\bar{P}x^{**} + \bar{Q}y^{**}), \end{cases} \tag{33}$$

where $z^{**}(0) = exp(T\Delta t)z_0$. We then have easily

$$z_{T-\chi}(\Delta t) \sim K\, exp(S\Delta t)P\, exp(T\Delta t)z_0. \tag{34}$$

We proceed in the same way for the $(\chi - T)$, $(\chi - T - \chi)$, and $(T - \chi - T)$ splittings.

For the splitting (NTS) we must reduce the following ODE (see (12) and (24)):

$$\begin{cases} \frac{dx^{**}}{dt} = P\chi_1(\bar{P}x^{**} + \bar{Q}y^{**}) + P\Delta T \\ \varepsilon\frac{dy^{**}}{dt} = Cx^{**} - Dy^{**} + \varepsilon Q\chi_1(\bar{P}x^{**} + \bar{Q}y^{**}) \end{cases} \tag{35}$$

with

$$\Delta T = \frac{exp(T\Delta t) - I}{\Delta t}z_0 \qquad \text{and} \qquad z^{**}(0) = z_0.$$

We obtain straightforwardly the reduced model in the form

$$\frac{dx^{**}}{dt} = P\chi_1 Kx^{**} + P\Delta T, \qquad 0 = Cx^{**} - Dy^{**}, \tag{36}$$

which leads after some calculations to:

$$z_{NTS}(\Delta t) \sim K\, exp(S\Delta t)\left[P + \int_0^{\Delta t} exp(-tS)\, dt\, P\, \frac{exp(T\,\Delta t) - I}{\Delta t}\right] z_0 \tag{37}$$

by using the definition $S = P\,\chi_1 K$. We recover the desired formula. ■

In order to have a more elegant formulation of these models we introduce the square matrix:

$$\Pi = K\,P. \tag{38}$$

LEMMA 3.3 (Projection onto the reduced model). Π *is a projection matrix onto the reduced model. Moreover for any square matrix* R,

$$K\, exp(P\,R\,K)\,P = exp(\Pi R)\,\Pi. \tag{39}$$

Proof. Let us first notice that

$$\Pi^2 = K\,P\,K\,P = K(PK)P \tag{40}$$

with

$$P\,K = P(\bar{P} + \bar{Q}D^{-1}C) = I \tag{41}$$

by using $P\,\bar{P} = I$ and $P\,\bar{Q} = 0$.

Π is therefore a projection matrix. The projection is made onto the reduced model since

$$z = \bar{P}x + \bar{Q}y, \qquad Cx - Dy = 0 \Leftrightarrow z = K\,P\,z = \Pi z. \tag{42}$$

The second point is obtained by recurrence: for any $n \geq 0$,

$$K(P\,R\,K)^n P = (\Pi R)^n \Pi \tag{43}$$

which ends the proof. ■

LEMMA 3.4 (Reduced solutions). *The reduced solutions are then*

$$\begin{cases} z_{WS}(\Delta t) \sim exp(\Pi(\chi_1 + T)\Delta t)\,\Pi z_0 \\ z_{T-\chi}(\Delta t) \sim exp(\Pi\chi_1\Delta t)\,\Pi\, exp(T\,\Delta t) z_0 \\ z_{\chi-T}(\Delta t) \sim exp(T\,\Delta t)\, exp(\Pi\chi_1\Delta t)\,\Pi z_0 \\ z_{\chi-T-\chi}(\Delta t) \sim exp\left(\Pi\chi_1\frac{\Delta t}{2}\right)\Pi\ exp(T\,\Delta t)\, exp\left(\Pi\chi_1\frac{\Delta t}{2}\right)\Pi z_0 \\ z_{T-\chi-T}(\Delta t) \sim exp\left(T\,\frac{\Delta t}{2}\right) exp(\Pi\chi_1\Delta t)\,\Pi\, exp\left(T\,\frac{\Delta t}{2}\right) z_0 \\ z_{NTS}(\Delta t) \sim exp(\Pi\chi_1\Delta t)\left[\Pi + \int_0^{\Delta t} exp(-t\,\Pi\chi_1)\, dt\ \Pi\frac{exp(T\,\Delta t) - I}{\Delta t}\right] z_0. \end{cases} \tag{44}$$

Proof. The proof is straightforward with Lemmas 3.1, 3.2, and 3.3. ■

3.4. Local Errors for the Reduced Models

In order to compare these reduced models we now perform an asymptotic expansion in Δt as usual. For the splitting scheme α, we will write

$$le_\alpha = z_\alpha(\Delta t) - z_{WS}(\Delta t), \tag{45}$$

where $z_\alpha(\Delta t)$ and $z_{WS}(\Delta t)$ denote the dominant term (up to first-order in ε) for the reduced solutions computed, respectively, for the splitting scheme α and for the exact solution. We have then the following result:

LEMMA 3.5 (Local error for the reduced models).

$$\begin{cases} le_{T-\chi} = \Pi T(I-\Pi)z_0\Delta t + O(\Delta t^2) \\ le_{\chi-T} = (I-\Pi)T\Pi z_0\Delta t + O(\Delta t^2) \\ le_{\chi-T-\chi} = \Pi T(I-\Pi)T\Pi z_0\frac{\Delta t^2}{2} + O(\Delta t^3) \\ le_{T-\chi-T} = (T\Pi + \Pi T - 2\Pi T\Pi)z_0\frac{\Delta t}{2} + O(\Delta t^2) \\ le_{NTS} = \Pi T(I-\Pi)z_0\Delta t + O(\Delta t^2) \end{cases} \tag{46}$$

Proof. After some tedious calculations. ■

Let us notice that the accuracy seems to be poorer than in the nonstiff case (as the naive application of the usual analysis could indicate it).

The key point is however that some splitting schemes will preserve the structure of the reduced model, which allows then to improve the local order: that is to say that the computed solution for these schemes is actually onto the reduced model. We indeed recover second-order local error for $(\chi - T)$ if $(I-\Pi)T\Pi = 0$, which is a very strong condition on the operators Π and T (a sufficient condition is for instance that they commute). On the other hand, we recover second-order local error for $(T-\chi)$ if $(I-\Pi)z_0 = 0$, which is only a condition on the current value z_0. This is rather easy to meet for some schemes as the next lemma shows it.

LEMMA 3.6 (Conservation of the exact reduced structure). *The splitting schemes* $(T-\chi)$, *(NTS), and* $(\chi - T - \chi)$ *conserve the reduced manifold defined by the projection matrix* Π:

$$\Pi z_{T-\chi} = z_{T-\chi}, \qquad \Pi z_{\chi-T-\chi} = z_{\chi-T-\chi}, \qquad \Pi z_{NTS} = z_{NTS}, \tag{47}$$

which must be compared with

$$\Pi z_{WS} = z_{WS}. \tag{48}$$

• *The splitting schemes* $(\chi - T)$ *and* $(T - \chi - T)$ *conserve the reduced structure under the commuting condition*

$$(I-\Pi)T\Pi = 0. \tag{49}$$

Proof. Let us consider a scheme α which can be put in the form

$$z_\alpha = exp(\Pi\, R')\Pi\, z'. \tag{50}$$

We have directly

$$\Pi exp(\Pi\, R') = \Pi - I + exp(\Pi R') \tag{51}$$

by using the asymptotic expansion of the exponential operator and the fact that Π is a projection matrix. Then

$$\Pi z_\alpha = exp(\Pi R')\Pi z' = z_\alpha \tag{52}$$

since $(I - \Pi)\Pi = 0$.
This concludes the proof for the schemes $(T - \chi)$, (*NTS*), and $(\chi - T - \chi)$.

- By using (44) we can put the schemes $(\chi - T)$ and $(T - \chi - T)$ in the form

$$z_\alpha = exp(\lambda T)\, exp(\Pi \chi_1 \Delta t)\Pi\, z', \qquad \lambda \in \mathbf{R}. \tag{53}$$

By using once more the asymptotic expansion of the exponential operator one has

$$z_\alpha = \Pi\, z' + (exp(\lambda T) - I)\Pi(*), \tag{54}$$

where the stars stand for uncomputed terms. Let us assume that the decoupling condition (49) is met:

$$T\Pi = \Pi T \Pi. \tag{55}$$

It is easy to obtain by recurrence that for any $n > 0$ one has

$$T^n \Pi = \Pi T^n \Pi, \tag{56}$$

which leads to

$$z_\alpha = \Pi\, z' + \Pi(exp(\lambda T) - I)\Pi(*). \tag{57}$$

This ensures $z_\alpha = \Pi\, z_\alpha$. ■

Remark [Commuting condition]. The terminology we use is justified by the equivalence of (49) with

$$(T\Pi - \Pi T)\Pi = 0. \tag{58}$$

The interpretation of this lemma is rather simple: the exact solution *WS* and the splitting schemes for which the stiff operator χ ends the iteration always preserve the reduced structure of the coupled system. Indeed the stiff operator alone determines this structure [21, 23, 28] whatever the nonstiff operator is and acts as a projection onto the reduced

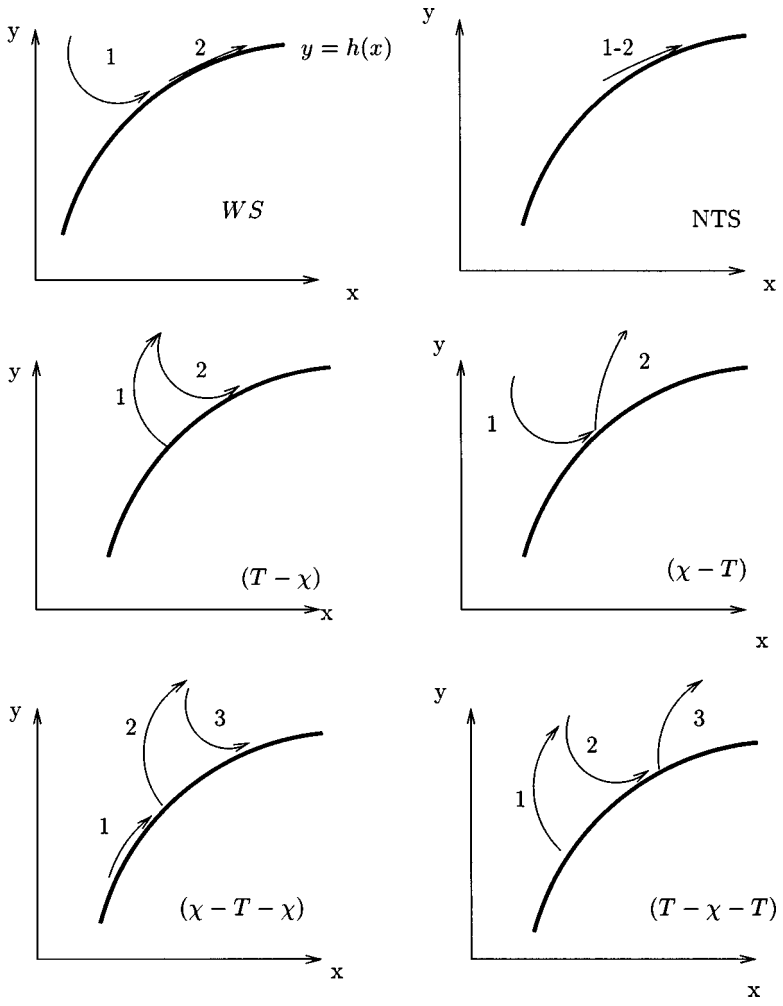

FIG. 2. Dynamical behavior of splitting schemes.

model. The integration of the nonstiff operator does not ensure that the solution remains near the reduced model unless the commuting condition is met.

The dynamical behavior of the exact solution *WS* and of the splitting schemes is indicated in Fig. 2. The trajectories are plotted in the phase space (x, y) and parametrized by time t. The wide black curve is the reduced algebraic constraint defined in the general case by $y = h(x)$ (in the linear case: $y = D^{-1}Cx$). The numbers 1, 2, and 3 stand for the substeps of the splitting schemes. Numbers 1 and 2 stand, respectively, for the inner and outer layers for the exact solution.

It is therefore logical to consider an initial condition z_0 belonging to the reduced manifold

$$\Pi z_0 = z_0$$

for the splitting methods conserving the reduced structure. Let us notice that the exact solution satisfies such a requirement after the transient phase (whose length is $O(\varepsilon)$). Under this assumption, which is therefore satisfied without any loss of generality, the local errors can be simplified.

LEMMA 3.7 (Local errors in the outer phase).

• *Under the condition* $\Pi z_0 = z_0$, *the local errors for the splitting schemes conserving the reduced structure are given by*

$$\begin{cases} le_{T-\chi} = O(\Delta t^2) \\ le_{\chi-T-\chi} = \Pi\, T(I-\Pi)T\,\Pi z_0 \frac{\Delta t^2}{2} + O(\Delta t^3) \\ le_{NTS} = O(\Delta t^2). \end{cases} \tag{59}$$

The splitting methods $(T-\chi)$ *and* (NTS) *are indeed first-order schemes while Strang splitting* $(\chi - T - \chi)$ *is only a first-order scheme.*

• *Strang splitting* $(\chi - T - \chi)$ *is a second-order method if and only if the commuting condition*

$$\Pi\, T(I-\Pi)T\,\Pi = 0 \tag{60}$$

is met.

• *We recover the usual order for the splitting schemes* $(\chi - T)$ *and* $(T - \chi - T)$ *under the commuting condition* (49).

Proof. Straightforwardly with $(I-\Pi)\Pi = 0$. ■

Remark [Commuting conditions]. The same remark as before holds for (60) written in the form

$$\Pi T(\Pi T - T\Pi) = 0. \tag{61}$$

Let us notice that the decoupling condition (49) implies (60).

Remark [Order reduction for ODEs]. The same phenomenon has already been observed for the numerical integration of stiff ODEs. In a few words, the study of local errors is usually made with a fixed stiffness ratio (that is to say a fixed Lipschitz constant) and a time step tending to 0; that is, the study is done for stiff systems in a framework of an explicit integration! The practical order of accuracy is unfortunately often lower than the theoretical one given by such an analysis. We refer to [20] and [4] for an alternative analysis. Let us mention that reduced solutions are also used in this context (see for instance [16, 18]). The use of such tools provided by numerical analysis is the topics of a joint work with Jan Verwer [33].

3.5. Errors for Slow and Fast Variables

It could be interesting to study the errors as a function of the dynamical behavior of the variables: are there any differences between slow variables and fast variables?

This is a relevant question in practice for at least two reasons:

• one is not necessary interested in having good accuracy for all variables (e.g., radicals may be not followed in chemical kinetics),

• such a study could indicate that it is better to work in a basis different from the initial one.

We first investigate the case of purely slow variables (defined as the variables not concerned by stiffness). We investigate thereafter the case of the partitioned basis as defined in Section 3.2.

3.5.1. *Purely Slow Variables*

Let us define the purely slow variables as those corresponding to null lines of χ_0. Such purely slow variables are not directly affected by stiffness. It may then be expected that accuracy is not greatly degraded for them.

LEMMA 3.8 (Splitting errors for purely slow variables). *The local error for purely slow variables is of second-order for the splitting scheme* $(\chi - T)$.

Proof. We are going to investigate the local error derived in Lemma 3.5. Let us recall that

$$le_{\chi-T} = (I - \Pi)T\Pi z_0 \Delta t + O(\Delta t^2). \tag{62}$$

Let i be the number of purely slow variables (eventually zero). Let us suppose that (eventually after a permutation) the first variables are purely slow variables. We have then by keeping the same notations as before

$$P = \begin{bmatrix} I & 0 \\ 0 & a \end{bmatrix}, \tag{63}$$

where I is the $i \times i$ identity matrix and a is a $(n - p - i) \times (n - i)$ matrix. Moreover

$$Q = [0 \quad b], \tag{64}$$

where b is a $p \times (n - i)$ matrix. We can then find $\bar{P}$ and $\bar{Q}$ in the form

$$\bar{P} = \begin{bmatrix} I & 0 \\ 0 & \bar{a} \end{bmatrix}, \qquad \bar{Q} = \begin{bmatrix} 0 \\ \bar{b} \end{bmatrix}, \tag{65}$$

where I is the $i \times i$ identity matrix, $\bar{a}$ is a $(n - i) \times (n - p - i)$ matrix and $\bar{b}$ is a $(n - i) \times p$ matrix.

Let us partition the $p \times (n - p)$ matrix C in the form

$$C = [C_1 \quad C_2] \tag{66}$$

with C_1 and C_2 being, respectively, $p \times i$ and $p \times (n - p - i)$ matrices. We can then easily compute

$$K = \bar{P} + \bar{Q}D^{-1}C = \begin{bmatrix} I & 0 \\ \bar{b}D^{-1}C_1 & \bar{a} + \bar{b}D^{-1}C_2 \end{bmatrix} \tag{67}$$

in the same block partitioning.

In the same way

$$\Pi = KP = \begin{bmatrix} I & 0 \\ * & * \end{bmatrix}, \qquad I - \Pi = \begin{bmatrix} 0 & 0 \\ * & * \end{bmatrix}, \tag{68}$$

where the asterisks stand for uncomputed blocks. We conclude with

$$(I - \Pi)T = \begin{bmatrix} 0 & 0 \\ * & * \end{bmatrix}, \tag{69}$$

which concludes the proof by using (62). ∎

3.5.2. *Partitioned Basis*

We now investigate the choice of the basis: can we find a set of variables (other than purely slow variables) such that the splitting error is lowered? We will study the error in the partitioned basis.

For convenience we assume that

$$P = [I \quad 0], \qquad Q = [0 \quad I], \qquad z = [x \quad y]^t. \tag{70}$$

This can be done without any loss of generality by working directly in the partitioned basis (x, y) as defined in Section 3.2. We then have in this basis (with a coherent block partitioning)

$$\Pi = \begin{bmatrix} I & 0 \\ D^{-1}C & 0 \end{bmatrix}, \tag{71}$$

and we will write

$$T = \begin{bmatrix} T_{xx} & T_{xy} \\ T_{yx} & T_{yy} \end{bmatrix}. \tag{72}$$

LEMMA 3.9 (Local errors in the partitioned basis). *In the partitioned basis,*

$$\begin{cases} le_{\chi-T-\chi} = \begin{bmatrix} T_{xy}U & 0 \\ D^{-1}CT_{xy}U & 0 \end{bmatrix} z_0 \dfrac{\Delta t^2}{2} \\ le_{\chi-T} = \begin{bmatrix} 0 & 0 \\ U & 0 \end{bmatrix} z_0 \Delta t \\ le_{T-\chi-T} = \begin{bmatrix} -T_{xy}\, D^{-1}C & T_{xy} \\ U - D^{-1}CT_{xy}D^{-1}C & D^{-1}CT_{xy} \end{bmatrix} z_0 \dfrac{\Delta t}{2} \end{cases} \tag{73}$$

with

$$U = T_{yx} + T_{yy}D^{-1}C - D^{-1}C(T_{xx} + T_{xy}D^{-1}C). \tag{74}$$

Proof. The proof is easy to perform by using Lemma 3.5 and by computing the matrices. ■

Remark [Lumped species and operator splitting]. Let us notice that some interesting simplifications occur if $T_{xy} = 0$. By remembering that T stands for the matrix of diffusive coefficients in the framework of Reaction-Diffusion equations, this assumption is met if intermolecular diffusion is not taken into account (which is the case for turbulent diffusion).

The application of the previous results indicates then the loss of accuracy for the splitting schemes $(\chi - T)$ and $(T - \chi - T)$ is associated only with the fast variables.

Lumped species [26, 27] are widely used in applications such as Air Pollution Modeling. This trick is equivalent to changing the basis of variables and with working directly with slow species x (in our notation). Lumped species are recommended for use in tailor-made numerical solvers based on reduced mechanisms [6, 26]. The former result is a confirmation that it is actually always better (whatever the numerical solver is) to use lumped species in the framework of operator splitting: the accuracy for lumped species is indeed better than for pure fast species if the splitting schemes $(\chi - T)$ and $(T - \chi - T)$ are used.

3.6. Well-Partitioned Time Scales

Let us now consider the case for which χ is a purely fast operator while T is a slow operator. At the algebraic level one then has

$$\chi_1 = 0. \tag{75}$$

This can always be obtained in the general case by modifying the splitting method and by adding χ_1 to T before splitting.

It would be interesting to recover the intuitive feeling that the better partitioned the time scales are, the more accurate is the splitting scheme.

3.6.1. *Singular Perturbation as a Time Splitting Approach in the Well-Partitioned Case*

The former analysis holds but the comparison of the exact solution *WS* with the splitting $(\chi - T)$ is enlightening. The singular perturbation theory indeed justifies that the initial slow–fast model can be approximated up to some orders in ε by integrating successively the fast and then the slow operator. The scheme $(\chi - T)$ then seems to be highly natural. Why is there a loss of accuracy even in this well-partitioned case ? Let us recall the singular perturbation procedure by formulating Theorem 3.1 in the following way:

1. First integrate the inner layer on $[0, \infty[$:

$$\frac{dx^*}{d\tau} = 0, \qquad \frac{dy^*}{d\tau} = Cx^* - Dy^*, \qquad \text{where } \tau = \frac{t}{\varepsilon}, \tag{76}$$

which provide the modified initial conditions (27). This is exactly the first step of the splitting scheme $(\chi - T)$ as $\frac{\Delta t}{\varepsilon}$ tends to $+\infty$!

2. Then integrate the outer layer on $[0, \Delta t]$:

$$\frac{dz^{**}}{dt} = \Pi T z^{**}, \tag{77}$$

where the projection matrix has to be applied in order to modify the initial term T. This is exactly the reduced model (26). This must be compared with the second step of the splitting scheme $(\chi - T)$:

$$\frac{dz^{**}}{dt} = T z^{**}. \tag{78}$$

The error for this scheme is obviously associated with the departure from the reduced manifold as it has been already proved in the former results. The most accurate splitting scheme would be the scheme derived from singular perturbation theory, that we could (formally) write $(\chi - \Pi T)$. Let us notice that under the assumption $\Delta t \gg \varepsilon$, the accuracy of such a scheme is dominated by ε and does not depend on the splitting timestep Δt in this sense!

Remark [Reduction vs operator splitting]. There is a kind of *trade-off* between Operator Splitting and Reduction. The most physical splitting (*fast–slow*) is given by the reducing approach (that is to say, the singular perturbation procedure): this is a highly accurate splitting (the error does not depend on Δt) but it is an expensive method (one must compute the projection matrix Π: that is to say, to compute the reduced chemical mechanism in

TABLE I
Reduction versus Operator Splitting

Method	CPU performance	Accuracy
Reduction	could be expensive (compute $\Pi(z)$)	high: $O(\varepsilon)$
Splitting slow–fast	good	medium: $O(\Delta t)$
Splitting fast–slow	good	low

the usual framework [26]). Slow–fast operator splitting $(T-\chi)$ is then the only alternative method: it is a very cheap one (nothing to do) but the price is the splitting error due to the fact that it is a nonphysical splitting (see Table I).

3.6.2. *A Particular Case: Highly Accurate Splitting*

Such an attractive feature can be performed with the classical splitting schemes $(T-\chi)$, (NTS), and $(\chi - T - \chi)$ in particular cases.

LEMMA 3.10 (Commuting case). *Under the commuting condition*

$$\Pi T(I-\Pi) = 0 \tag{79}$$

the splitting schemes $(T-\chi)$, (NTS), *and* $(\chi - T - \chi)$ *are highly accurate in the coarse case* $(\Delta t \gg \varepsilon)$:

$$le_{T-\chi} = O(\varepsilon), \qquad le_{\chi-T-\chi} = O(\varepsilon), \qquad le_{NTS} = O(\varepsilon). \tag{80}$$

Proof. Let us recall that the reduced solutions, which have been previously computed, are approximations of the exact solutions associated with each scheme up to first order in ε:

$$\begin{cases} z_{WS} \sim exp(\Pi\, T\, \Delta t)\Pi z_0 \\ z_{T-\chi} \sim \Pi exp(T\,\Delta t) z_0 \\ z_{\chi-T-\chi} \sim \Pi exp(T\,\Delta t)\Pi z_0 \\ z_{NTS} \sim \Pi exp(T\,\Delta t) z_0. \end{cases} \tag{81}$$

We use asymptotic expansions of the exponential operators and the proof is then straightforward by recurrence. Let us suppose that for any $n > 1$,

$$(\Pi\, T)^n = \Pi\, T^n, \qquad \Pi T^n \Pi = \Pi T^n. \tag{82}$$

Under the assumption $\Pi T = \Pi T \Pi$

$$(\Pi T)^{n+1} = (\Pi T)(\Pi T)^n = (\Pi T)\Pi T^n = (\Pi T \Pi) T^n = \Pi T\, T^n \tag{83}$$

$$\Pi T^{n+1}\Pi = \Pi T \Pi T^n \Pi = \Pi T(\Pi T^n \Pi) = \Pi T \Pi T^n = \Pi T\, T^n, \tag{84}$$

which concludes the proof. ∎

Remark [Commuting condition]. Equation (79) can be written as

$$\Pi(T\Pi - T\Pi) = 0. \tag{85}$$

This is equivalent to $T_{xy} = 0$ (with the same notations as before) since

$$\Pi\, T(I - \Pi) = \begin{bmatrix} -T_{xy}D^{-1}C & T_{xy} \\ -D^{-1}CT_{xy}D^{-1}C & D^{-1}CT_{xy} \end{bmatrix}. \tag{86}$$

It is therefore associated with the following form of system to be split:

$$\frac{dx}{dt} = T_{xx}x, \qquad \varepsilon\frac{dy}{dt} = Cx - Dy + \varepsilon(T_{yx}x + T_{yy}y). \tag{87}$$

Let us notice that the usual analysis does not give any such information. A direct calculation leads for instance to a commutator:

$$\chi T - T\chi = \begin{bmatrix} 0 & 0 \\ CT_{xx} - DT_{xy} - T_{xy}C & T_{yy}D - DT_{yy} \end{bmatrix}. \tag{88}$$

Remark [Validity of the assumption]. Let us notice that such a condition may occur in the Reaction-Diffusion case with a purely fast chemistry and a diagonal diffusion matrix. Formally,

$$T_{xx} = k_{xx}I, \qquad T_{xy} = T_{yx} = 0, \qquad T_{yy} = k_{yy}I, \tag{89}$$

where I stands for Identity matrices.

This is the case for instance in Air Pollution Modeling where the turbulent diffusion matrix is a diagonal matrix (no diffusion between species). We will go back to this remark in Section 5.

Remark [Decoupling condition]. Let us notice that the decoupling condition (79) (which ensures highly accurate splitting) implies the decoupling condition (60) (which ensures that $(\chi - T - \chi)$ is a second-order method).

3.7. Summary of the Main Results

Let us now summarize the main points of our analysis in the coarse case:

- *The "first-order" splitting schemes* $(T - \chi)$ *and* (NTS) *are indeed first-order schemes.*
- *The "second-order" splitting scheme* $(\chi - T - \chi)$ *is only a first-order scheme in the general case.*
- *The respectively "first-order" and "second-order" schemes* $(\chi - T)$ *and* $(T - \chi - T)$ *give only poor accuracy. However, for purely slow variables we recover the usual orders.*
- *The usual orders are maintained under the condition*

$$(I - \Pi)T\Pi = 0. \tag{90}$$

Remark [After all, stiffness is a good thing!]. What about the global error as order reduction occurs?

The key point is that stiffness has a stabilizing effect and that the local errors for fast species do not propagate. The following simple example is an enlightening illustration (the general case can be computed in the same way),

$$\frac{dx}{dt} = -x, \qquad \frac{dy}{dt} = \frac{x - y}{\varepsilon}. \tag{91}$$

The slow and fast operators are repectively associated with the time integration of x and y. Let n be the iteration index. The exact solution is then

$$x_{n+1}^{WS} = e^{-\Delta t} x_n, \qquad y_{n+1}^{WS} = \frac{x_n e^{-\Delta t}}{1-\varepsilon} + \left(y_n - \frac{x_n}{1-\varepsilon} \right) exp\left(-\frac{\Delta t}{\varepsilon} \right). \tag{92}$$

Let $(x_n + \eta_n^1, y_n + \eta_n^2)$ be the solution computed with the bad splitting (fast–slow), where η_n^1 and η_n^2 denote global errors after n iterations. We have very easily for the splitting fast–slow:

$$x_{n+1} = e^{-\Delta t}\left(x_n + \eta_n^1\right), \qquad y_{n+1} = x_n + \eta_n^1 + \left(y_n + \eta_n^2 - x_n - \eta_n^1\right) e^{-\frac{\Delta t}{\varepsilon}}. \tag{93}$$

The interesting point is that the error for slow species (η_n^1) is stabilized and that the first-order local error for fast species (η_n^2) has been put to zero (since it appears in the boundary layer term)! The only error which propagates is the error for slow species (η_n^1) which is a second-order error.

4. SOME NUMERICAL TESTS IN THE LINEAR CASE

We have performed some numerical tests in order to illustrate the previous analysis in the linear case. We have used the solver LSODE [7] with highly accurate absolute and relative error parameters: the parameters *rtol* and *atol* are set, respectively, to 10^{-10} and 10^{-15}.

We have integrated linear ODEs in $\mathbf{R}^3$ and we write as (u, v, w) the variables to be integrated.

In the following, we have mainly studied a fast part of the stiff operator given by

$$\frac{\chi_0}{\varepsilon} = \begin{bmatrix} -1000 & 1000 & 0 \\ 1000 & -1000 & 0 \\ 0 & 0 & 0 \end{bmatrix}, \tag{94}$$

which corresponds to a stiffness ratio $\varepsilon = 10^{-3}$. The following results are not modified by changing ε.

It is easy to derive the projection matrix

$$\Pi = \frac{1}{2} \begin{bmatrix} 1 & 1 & 0 \\ 1 & 1 & 0 \\ 0 & 0 & 2 \end{bmatrix}. \tag{95}$$

In the previous terminology u and v are concerned with fast dynamics while w is a purely slow variable.

We have used a splitting timestep Δt ranging from 1. to 10^{-3}. We expect to recover the usual analysis with $\Delta t = 10^{-3}$ and to confirm our analysis for the coarse case $\Delta t = 0.1$ and $\Delta t = 0.01$. The case $\Delta t = 1.$ is highly coarse (even if it could correspond to the practical situation) and we expect only to confirm a *trend* in this case.

4.1. Nonstiff Case (Case 1)

We first study the nonstiff case with $\chi_0 = 0$ and

$$\chi = \chi_1 = \begin{bmatrix} -1 & 1 & 0 \\ 1 & -1 & 0 \\ 0 & 0 & 0 \end{bmatrix}, \qquad T = \begin{bmatrix} -3 & 1 & 0 \\ 2 & -2 & 0 \\ 0.2 & 0 & -1 \end{bmatrix}. \tag{96}$$

TABLE II

Case 1. Relative Error at $t = 4$

Δt	$(\chi - T)$	$(T - \chi)$	$(\chi - T - \chi)$	$(T - \chi - T)$
10^{-3}	$1.39 \cdot 10^{-4}$	$1.41 \cdot 10^{-4}$	~ 0	~ 0
10^{-2}	$1.39 \cdot 10^{-3}$	$1.39 \cdot 10^{-3}$	$1.21 \cdot 10^{-5}$	$7.6 \cdot 10^{-6}$
10^{-1}	$1.37 \cdot 10^{-2}$	$1.41 \cdot 10^{-2}$	$1.01 \cdot 10^{-3}$	$8.66 \cdot 10^{-4}$
1.	$8.8 \cdot 10^{-2}$	$11.4 \cdot 10^{-2}$	$6.3 \cdot 10^{-2}$	$5.14 \cdot 10^{-2}$

As Table II shows, we recover the classical behavior as Δt tends to 0. The order is actually confirmed and there is no difference between the first-order schemes (respectively, between the second-order schemes).

4.2. Stiff Case: $\chi_1 = 0$ and No Commuting Condition (Case 2)

We have kept the same matrix T as before but we have now

$$\chi = \frac{\chi_0}{\varepsilon} = \begin{bmatrix} -1000 & 1000 & 0 \\ 1000 & -1000 & 0 \\ 0 & 0 & 0 \end{bmatrix}, \tag{97}$$

which corresponds to the stiff case with $\chi_1 = 0$. Table III indicates that the schemes $(T - \chi)$ and $(\chi - T - \chi)$ have a better accuracy in the coarse case. $(\chi - T - \chi)$ is however only a first-order scheme in this regime (which is equivalent with $(T - \chi)$ in this particular case).

It is interesting to confirm the previous analysis concerning purely slow variables. In this case u and v are concerned with fast dynamics while w is a purely slow variable in the previous terminology. The relative errors plotted in Tables IV, V, VI and VII confirm that w is not affected by the loss of accuracy in the schemes $(\chi - T)$ and $(T - \chi - T)$ in contrast to fast variables u and v. The error for w is indeed the same for all schemes.

4.3. Stiff Case: $\chi_1 \neq 0$ and No Decoupling Condition (Case 3)

We have kept the same matrix T as before but we have now

$$\chi = \begin{bmatrix} -1000 & 1000 & 1 \\ 1000 & -1000 & 2 \\ 0 & 1 & -1 \end{bmatrix}, \tag{98}$$

which corresponds to $\chi_1 \neq 0$.

TABLE III

Case 2. Relative Error at $t = 1$

Δt	$(\chi - T)$	$(T - \chi)$	$(\chi - T - \chi)$	$(T - \chi - T)$
10^{-3}	$4.4 \cdot 10^{-4}$	$2.33 \cdot 10^{-4}$	$5.4 \cdot 10^{-5}$	$1.1 \cdot 10^{-4}$
10^{-2}	$6.4 \cdot 10^{-3}$	$4.88 \cdot 10^{-4}$	$4.87 \cdot 10^{-4}$	$3.13 \cdot 10^{-3}$
10^{-1}	$5.9 \cdot 10^{-2}$	$1.91 \cdot 10^{-3}$	$1.91 \cdot 10^{-3}$	$3.21 \cdot 10^{-2}$
1.	$21.8 \cdot 10^{-2}$	$8.31 \cdot 10^{-3}$	$8.31 \cdot 10^{-3}$	$17.8 \cdot 10^{-2}$

TABLE IV

Case 2. Relative Error at $t = 1$ for $(\chi - T)$

Δt	u	v	w
10^{-3}	$6.55 \cdot 10^{-4}$	$6.55 \cdot 10^{-4}$	$1.6 \cdot 10^{-5}$
10^{-2}	$9.35 \cdot 10^{-3}$	$9.35 \cdot 10^{-3}$	$4.7 \cdot 10^{-4}$
10^{-1}	$8.6 \cdot 10^{-2}$	$8.6 \cdot 10^{-2}$	$3.0 \cdot 10^{-3}$
1.	$32.0 \cdot 10^{-2}$	$32.0 \cdot 10^{-2}$	$2.4 \cdot 10^{-2}$

TABLE V

Case 2. Relative Error at $t = 1$ for $(T - \chi - T)$

Δt	u	v	w
10^{-3}	$1.6 \cdot 10^{-4}$	$1.6 \cdot 10^{-4}$	$1.6 \cdot 10^{-5}$
10^{-2}	$4.5 \cdot 10^{-3}$	$4.5 \cdot 10^{-3}$	$4.6 \cdot 10^{-4}$
10^{-1}	$4.6 \cdot 10^{-2}$	$4.6 \cdot 10^{-2}$	$4.5 \cdot 10^{-3}$
1.	$26.0 \cdot 10^{-2}$	$26.0 \cdot 10^{-2}$	$1.7 \cdot 10^{-2}$

TABLE VI

Case 2. Relative Error at $t = 1$ for $(T - \chi)$

Δt	u	v	w
10^{-3}	$3.4 \cdot 10^{-4}$	$3.4 \cdot 10^{-4}$	$1.6 \cdot 10^{-5}$
10^{-2}	$5.0 \cdot 10^{-4}$	$5.0 \cdot 10^{-4}$	$4.7 \cdot 10^{-4}$
10^{-1}	$5.0 \cdot 10^{-4}$	$5.0 \cdot 10^{-4}$	$4.7 \cdot 10^{-3}$
1.	$5.0 \cdot 10^{-4}$	$5.0 \cdot 10^{-4}$	$2.4 \cdot 10^{-2}$

TABLE VII

Case 2. Relative Error at $t = 1$ for $(\chi - T - \chi)$

Δt	u	v	w
10^{-3}	$7.3 \cdot 10^{-5}$	$7.3 \cdot 10^{-5}$	$1.5 \cdot 10^{-5}$
10^{-2}	$5.0 \cdot 10^{-4}$	$5.0 \cdot 10^{-4}$	$4.7 \cdot 10^{-4}$
10^{-1}	$5.0 \cdot 10^{-4}$	$5.0 \cdot 10^{-4}$	$4.7 \cdot 10^{-3}$
1.	$5.0 \cdot 10^{-4}$	$5.0 \cdot 10^{-4}$	$2.4 \cdot 10^{-2}$

TABLE VIII

Case 3. Relative Error at $t = 4$

Δt	$(\chi - T)$	$(T - \chi)$	$(\chi - T - \chi)$	$(T - \chi - T)$
10^{-3}	$4.5 \cdot 10^{-4}$	$3.0 \cdot 10^{-4}$	$7.5 \cdot 10^{-5}$	$1.3 \cdot 10^{-4}$
10^{-2}	$6.5 \cdot 10^{-3}$	$2.0 \cdot 10^{-3}$	$1.8 \cdot 10^{-3}$	$3.7 \cdot 10^{-3}$
10^{-1}	$5.9 \cdot 10^{-2}$	$2.0 \cdot 10^{-2}$	$1.8 \cdot 10^{-2}$	$3.7 \cdot 10^{-2}$
1.	$18.3 \cdot 10^{-2}$	$11.4 \cdot 10^{-2}$	$9.2 \cdot 10^{-2}$	$17.9 \cdot 10^{-2}$

The global results in Table VIII confirm that the splitting $(T-\chi)$ and $(\chi-T-\chi)$ are to be preferred in the coarse case and that $(\chi-T-\chi)$ is only a first-order scheme.

Let us study in particular the errors associated with fast variables (u and v) and purely slow ones (w). The previous theoretical study (see Lemma 3.5) led to

$$\begin{cases} le_{\chi-T} = (I-\Pi)T\Pi z_0 \Delta t + O(\Delta t^2) \\ le_{T-\chi-T} = (T\Pi + \Pi T - 2\Pi T\Pi) z_0 \frac{\Delta t}{2} + O(\Delta t^2). \end{cases} \tag{99}$$

We obtain straightforwardly

$$(I-\Pi)T\Pi = \frac{1}{2}\begin{bmatrix} -1 & -1 & 0 \\ 1 & 1 & 0 \\ 0 & 0 & 0 \end{bmatrix} \tag{100}$$

and

$$T\Pi + \Pi T - 2\Pi T\Pi = \frac{1}{4}\begin{bmatrix} -1 & -1 & 0 \\ 1 & 1 & 0 \\ 0.2 & -0.2 & 0 \end{bmatrix}. \tag{101}$$

The splitting $(\chi - T)$ then admits a low error for w in contrast to the splitting $(T-\chi-T)$. For the variables u and v, one must verify

$$le_{T-\chi-T} \sim \frac{1}{2} le_{\chi-T}. \tag{102}$$

This is confirmed by Tables IX and X which indicate that the splitting $(T-\chi-T)$ performs better than the splitting $(\chi-T)$ in the coarse case.

In the same way the splitting $(T-\chi)$ and $(\chi-T-\chi)$ have a better accuracy for coarse splitting time step. The splitting $(\chi-T-\chi)$ is however only a first-order scheme (see Tables XI and XII).

4.4. Stiff Case: $\chi_1 \neq 0$ and Commuting Condition (Case 4)

We keep the same matrix χ as before. The matrix T is now such that $(I-\Pi)T\Pi = 0$ is met. This is a sufficient condition for recovering the usual order: see equations (49) and (60). We take

$$T = \begin{bmatrix} -3 & 1 & 0 \\ -3 & 1 & 0 \\ 0.2 & 0 & -1 \end{bmatrix}. \tag{103}$$

Table XIII confirms the theoretical analysis. The splitting schemes indeed recover their usual orders. The sequential order of operators to be split has lost any influence on the accuracy.

5. THE REACTION-DIFFUSION CASE: SOME EXAMPLES

We will now focus on some examples of coupling between chemistry and diffusion in a monodimensional case. One field of application is Air Pollution Modeling. The main errors

TABLE IX

Case 3. Relative Error at $t = 4$ for $(\chi - T)$

Δt	u	v	w
10^{-3}	$7.5 \cdot 10^{-4}$	$5.6 \cdot 10^{-4}$	$5.1 \cdot 10^{-5}$
10^{-2}	$1.1 \cdot 10^{-2}$	$7.3 \cdot 10^{-3}$	$8.9 \cdot 10^{-4}$
10^{-1}	$10.4 \cdot 10^{-2}$	$6.4 \cdot 10^{-2}$	$9.4 \cdot 10^{-3}$
1.	$40.5 \cdot 10^{-2}$	$14.5 \cdot 10^{-2}$	$4.1 \cdot 10^{-4}$

TABLE X

Case 3. Relative Error at $t = 4$ for $(T - \chi - T)$

Δt	u	v	w
10^{-3}	$2.1 \cdot 10^{-4}$	$1.0 \cdot 10^{-4}$	$7.6 \cdot 10^{-5}$
10^{-2}	$6.1 \cdot 10^{-3}$	$2.8 \cdot 10^{-3}$	$2.2 \cdot 10^{-3}$
10^{-1}	$6.2 \cdot 10^{-2}$	$2.8 \cdot 10^{-2}$	$2.1 \cdot 10^{-2}$
1.	$33.6 \cdot 10^{-2}$	$12.6 \cdot 10^{-2}$	$7.3 \cdot 10^{-2}$

TABLE XI

Case 3. Relative Error at $t = 4$ for $(T - \chi)$

Δt	u	v	w
10^{-3}	$3.2 \cdot 10^{-4}$	$3.7 \cdot 10^{-4}$	$2.0 \cdot 10^{-4}$
10^{-2}	$8.2 \cdot 10^{-4}$	$1.8 \cdot 10^{-3}$	$3.4 \cdot 10^{-3}$
10^{-1}	$1.3 \cdot 10^{-2}$	$1.4 \cdot 10^{-2}$	$3.3 \cdot 10^{-2}$
1.	$8.3 \cdot 10^{-2}$	$8.4 \cdot 10^{-2}$	$17.5 \cdot 10^{-2}$

TABLE XII

Case 3. Relative Error at $t = 4$ for $(\chi - T - \chi)$

Δt	u	v	w
10^{-3}	$1.7 \cdot 10^{-5}$	$1.3 \cdot 10^{-4}$	$7.6 \cdot 10^{-5}$
10^{-2}	$1.2 \cdot 10^{-3}$	$2.2 \cdot 10^{-3}$	$2.2 \cdot 10^{-3}$
10^{-1}	$1.6 \cdot 10^{-2}$	$1.7 \cdot 10^{-2}$	$2.2 \cdot 10^{-2}$
1.	$7.6 \cdot 10^{-2}$	$7.7 \cdot 10^{-2}$	$12.3 \cdot 10^{-2}$

TABLE XIII

Case 4. Relative Error at $t = 4$

Δt	$(\chi - T)$	$(T - \chi)$	$(\chi - T - \chi)$	$(T - \chi - T)$
10^{-3}	$3.25 \cdot 10^{-4}$	$3.26 \cdot 10^{-4}$	~ 0	~ 0
10^{-2}	$3.25 \cdot 10^{-3}$	$3.26 \cdot 10^{-3}$	$2.88 \cdot 10^{-5}$	$2.63 \cdot 10^{-5}$
10^{-1}	$3.26 \cdot 10^{-2}$	$3.26 \cdot 10^{-2}$	$2.35 \cdot 10^{-3}$	$2.01 \cdot 10^{-3}$
1.	$39.0 \cdot 10^{-2}$	$44.8 \cdot 10^{-2}$	$21.5 \cdot 10^{-2}$	$17.8 \cdot 10^{-2}$

induced by operator splitting are indeed produced by the uncoupling between chemistry and diffusion (see [13] for a systematic study) and by the treatment of Boundary Conditions. We do not deal with this last point.

Let us mention that the previous theoretical analysis must be applied carefully since the chemical part is nonlinear. The first example below proves that the key point of the analysis is the two timescales structure and does not rely on the linearity.

In the sequel we compute relative errors in the following way: a reference solution (z_{WS}) is computed with the Method of Lines. If z_α is the solution given by a splitting method, we define the usual L_2 error norms as

$$ERR_2^i(n,m) = \frac{z_{MOL}^i(n,m) - z_\alpha^i(n,m)}{z_{MOL}^i(n,m) + atol} \tag{104}$$

$$R_2^i(n) = \sqrt{\frac{1}{H}\sum_{m=1}^{m=M} ERR_2^i(n,m)^2\, dx(m)}, \qquad R_2(n) = \frac{1}{I}\sum_{i=1}^{i=I} R_2^i(n), \tag{105}$$

where i, n, and m stand, respectively, for the species, iteration, and grid cell index (whose whole numbers are I, N, and M), $atol$ is a tolerance parameter, $dx(j)$ is the length of the j grid cell, and H is the vertical height ($\sim$1200 m).

There are 10 vertical cells (which is a realistic number). The turbulent diffusion coefficient is constant ($k = 5\mathrm{m}^2 \cdot \mathrm{s}^{-1}$).

Finally, let us mention that $\Delta t = 900$ s is the classical value advocated for the splitting time step [17].

5.1. Chapman's Cycle for Ozone and Slow Diffusion (Case 5)

Let us first study as a simple case the tropospheric cycle of Ozone with slow diffusion. The kinetic scheme is given in simplified form [10] by

$$NO + O_3 \xrightarrow{k_1} NO_2\,, \qquad NO_2 \xrightarrow{k_2} NO + O_3, \tag{106}$$

where k_1 and k_2 are the kinetic rates given by the law of mass action.

We assume that the chemical kinetics induces a stiff dynamical system and we write formally the chemical rate as

$$\frac{\Omega}{\varepsilon} = k_1(NO)O_3 - k_2 NO_2. \tag{107}$$

We now study the reaction-diffusion system given by

$$\frac{dNO}{dt} = -\frac{\Omega}{\varepsilon} + k\Delta NO, \qquad \frac{dNO_2}{dt} = -\frac{\Omega}{\varepsilon} + k\Delta NO_2, \qquad \frac{dO_3}{dt} = \frac{\Omega}{\varepsilon} + k\Delta O_3, \tag{108}$$

where k is a fixed turbulent diffusity coefficient (let us say $k \sim O(1)$) and Δ denotes the Laplacian operator (or a spatial discretization by finite differences). We do not take into account any boundary conditions in this formal study.

By introducing the usual lumped species [6, 26, 27]

$$NO_x = NO + NO_2, \qquad O_x = O_3 + NO_2, \tag{109}$$

TABLE XIV
Case 5. Relative Error R_2 at $t = 2000$

Δt	$(\chi - T)$	$(T - \chi)$
50	$3.3 \cdot 10^{-3}$	$2.0 \cdot 10^{-4}$
100	$6.5 \cdot 10^{-3}$	$1.0 \cdot 10^{-4}$
500	$3.45 \cdot 10^{-2}$	$1.8 \cdot 10^{-5}$
1000	$7.35 \cdot 10^{-2}$	$8.3 \cdot 10^{-6}$
2000	$16.6 \cdot 10^{-2}$	$3.0 \cdot 10^{-6}$

we have easily

$$\frac{dNO_x}{dt} = k\Delta NO_x, \qquad \frac{dO_x}{dt} = k\Delta O_x, \qquad \frac{dO_3}{dt} = \frac{\Omega}{\varepsilon} + k\Delta O_3. \tag{110}$$

If the current value is $(NO_x, O_x, O_3)_n$ (n being the iteration index), we have at the next splitting time step for the exact solution

$$(NO_x)_{n+1} = exp(\Delta\Delta t)(NO_x)_n, \qquad (O_x)_{n+1} = exp(\Delta\Delta t)(O_x)_n, \qquad \Omega_{n+1} \sim 0 \tag{111}$$

up to first order in ε, where Ω_{n+1} is the chemical rate Ω computed at iteration $n+1$.

The second substep of the splitting $(\Delta - \chi)$ is now defined by

$$\frac{dNO_x^{**}}{dt} = 0, \qquad \frac{dO_x^{**}}{dt} = 0, \qquad \frac{dO_3^{**}}{dt} = \frac{\Omega^{**}}{\varepsilon}, \tag{112}$$

where the initial conditions are the outputs of the first substep:

$$NO_x^{**}(0) = exp(\Delta\Delta t)(NO_x)_n, \qquad O_x^{**}(0) = exp(\Delta\Delta t)(O_x)_n \tag{113}$$

$$O_3^{**}(0) = exp(\Delta\Delta t)(O_3)_n \tag{114}$$

and we recover (111) and the solution WS up to first-order in ε. As mentioned before "the error does not depend on Δt in this coarse regime (with $\Delta t \gg \varepsilon$)"!

We have performed some sumerical tests with

$$k_1 = 1000, \qquad k_2 = 2000, \qquad k = 5 \tag{115}$$

in monodimensional computations. The results plotted in Table XIV confirm the previous analysis. The splitting $(T - \chi)$ is better as Δt increases due to the error for the transient

TABLE XV
Case 6. Final Relative Error R_2 at $t = 6 \times 3600$

Δt	$(\chi - T)$	$(T - \chi)$	$(\chi - T - \chi)$	$(T - \chi - T)$
90	$8.8 \cdot 10^{-3}$	$3.96 \cdot 10^{-3}$	$7.2 \cdot 10^{-4}$	$3.0 \cdot 10^{-3}$
300	$3.25 \cdot 10^{-2}$	$1.25 \cdot 10^{-2}$	$2.08 \cdot 10^{-3}$	$1.26 \cdot 10^{-2}$
900	$11.0 \cdot 10^{-2}$	$3.84 \cdot 10^{-2}$	$3.78 \cdot 10^{-3}$	$4.47 \cdot 10^{-2}$

TABLE XVI
Case 6. Global Relative Error

Δt	$(\chi - T)$	$(T - \chi)$	$(\chi - T - \chi)$	$(T - \chi - T)$
90	$16.0 \cdot 10^{-2}$	$1.74 \cdot 10^{-2}$	$1.06 \cdot 10^{-2}$	$5.0 \cdot 10^{-2}$
300	$110.0 \cdot 10^{-2}$	$2.58 \cdot 10^{-2}$	$1.55 \cdot 10^{-2}$	$33.0 \cdot 10^{-2}$
900	$189.0 \cdot 10^{-2}$	$5.9 \cdot 10^{-2}$	$1.27 \cdot 10^{-2}$	$63.0 \cdot 10^{-2}$

phase, which is given by

$$O\left(exp\left(-\frac{\Delta t}{\varepsilon}\right)\right). \tag{116}$$

This example is a good illustration of the stabilizing effect of stiffness for splitting schemes applied to well-partitioned systems.

5.2. A Real Case in Air Pollution Modeling (Case 6)

We have simulated a reaction-diffusion system with data given in [10]. Tables XV and XVI give, respectively, the L_2 relative error at the final time $t = T$ (the length of computation is 6 h) and L_2 integrated during the whole time integration (global error).

The boundary conditions are dry deposition and emissions at the ground and no flux at the top. We have performed a (crude) sensitivity analysis by canceling all boundary conditions. As it does not affect the previous results, we think that the observed phenomena do not have any connection with the treatment of boundary conditions.

We refer to [25] for an extensive numerical study in the framework of Air Pollution Modeling.

CONCLUSION

We have proved here that the usual analysis for splitting errors fails in the stiff case. The singular perturbation theory provides an appropriate framework for studying the coarse case met in practice.

The comparison of the asymptotic behavior of each splitting scheme leads one to advocate some of them: the key point is that the operator sequence is crucial and that the stiff operator has always to be last in the splitting process.

Such an analysis is confirmed by numerical tests for the linear case and by examples derived from air pollution modeling (Reaction-Diffusion).

In the future we will investigate the nonlinear case and the case of Reaction-Diffusion more deeply.

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