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Spectral approximation of solutions to the chemical master equation

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ABSTRACT

The master equation of chemical reactions is an accurate stochastic description of general systems in chemistry. For D reacting species this is a differential-difference equation in D dimensions, exactly soluble for very simple systems only.

We propose and analyze a novel solution strategy in the form of a Galerkin spectral method with a favorable choice of basis functions. A spectral approximation theory in the corresponding spaces is developed and the issue of stability is discussed.

The convergence properties of the method are demonstrated by the numerical solution of two model problems with known solutions and a third problem for which no solution is known. It is shown that the method is effective and accurate, providing a viable alternative to other solution methods when the dimensionality is not too high.

1. Introduction

The *Markov property* of stochastic processes plays an important role in many descriptions of real-world phenomena. For a physical system observed at discrete times $t_1 < t_2 < \cdots t_n$ it states that the probability for the observation (y_n, t_n) conditioned on the system's history satisfies

$$\Pr(y_n, t_n | y_1, t_1; \ldots; y_{n-1}, t_{n-1}) = \Pr(y_n, t_n | y_{n-1}, t_{n-1}), \tag{1.1}$$

i.e. that the future evolution depends on the present state only. Although (1.1) is not always fulfilled exactly, it is frequently a very accurate and useful approximation. In particular, the Markov assumption is accurate when the discrete time chosen is sufficiently coarse in comparison with the often very short auto-correlation time of the system. Systems satisfying (1.1) can be described using only the initial probability $\Pr(y_1, t_1)$ and the *transition probability function* $\Pr(y_s, s | y_t, t)$ [2].

The *master equation* is a consequence of the Markov property for a discrete state space in continuous time. In particular, if a chemical system of D reacting species is described by counting the number of molecules of each kind, then the master equation accurately governs the dynamics of the probability distribution for the system. In fact, one can show that the chemical master equation is *exact* under the conditions that the system is well-stirred and in thermal equilibrium [18].

The resulting description is a differential-difference equation in D dimensions and therefore suffers from the well-known "curse of dimensionality"; — each species adds one dimension to the problem leading to a computational complexity that grows exponentially. Only a few examples are analytically solvable, and effective numerical methods for solving the master equation are of both practical and theoretical interest.

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A common deterministic model is the *reaction-rate* equations. This is a set of D ordinary differential equations (ODEs) approximating the expected values of the concentrations of the species in the system. There are, however, many systems for which the reaction-rate equations fail to reproduce actual behavior [26]. For instance, biological systems inside living cells frequently consist of fewer than 10^2 molecules of some of the species [19] and stochastic effects are therefore more pronounced. Additionally, such systems are often driven towards critical points for various biological reasons. Close to such points, small random fluctuations in one variable may slowly "leak" probability mass in a direction that on a longer timescale drastically affects the rest of the system.

There are several stochastic simulation techniques that offer the ability to exactly follow sample trajectories of the master equation. *Gillespie's Stochastic Simulation Algorithm* (SSA) [16] is perhaps the most well-known of these methods. Although SSA is quite effective for many systems, models in molecular biology are often very stiff, and explicit simulation techniques should not be applied directly [5,9,20].

We mention also that there are analytical techniques for solving the master equation. Under certain assumptions *a priori*, the Ω-expansion [23, Ch. X] yields an asymptotic expansion of the solution in inverse powers of the size Ω of the system. Another analytical approach is the *Poisson representation* [15, Ch. 7.7], where the solution is written as a superposition of uncorrelated Poisson distributions.

Recent *numerical* considerations for the master equation include the numerical solution of the *Fokker–Planck equation* [14] and the adaption of the *Sparse grids technique* [21]. When viewed as a master equation for fairly general *continuous* stochastic processes [15, Ch. 7.2], numerical solution of the Fokker–Planck equation is an interesting subject in itself. As a continuous approximation to the master equation, however, it is difficult to say how good the approximation will be [23, Ch. VIII]. The sparse grids technique directly aims to reduce the computational complexity of high dimensional smooth problems. Its application to the master equation is quite recent and would appear promising.

In the present paper we will provide the foundation for a spectral method that applies directly to the master equation. For this purpose, theoretical considerations, including approximation properties and stability are developed here. For implementation issues, efficiency concerns and experiments on representative models, see [13]. Both papers are adapted from the earlier work [12].

The proposed spectral method is unusual in that the basis functions are orthogonal with respect to a *discrete* measure. This suits the discrete character of the solution and avoids the need for continuous approximations to the master operator. The method's efficiency thus depends on the spectral representation of smooth solutions over discrete sets, where "smooth" has to be defined in this new context. This does not completely remove the dimensional curse, but the spectral representation is more efficient than any direct representation. We note that our proposed scheme is reminiscent of an approach considered earlier in [7], and will further comment on this point in Section 4.

The rest of the paper is organized as follows. Section 2 is devoted to a theoretical study of the master equation and the suggested numerical solution method. We define the master equation and briefly touch upon some of its properties. Suitable approximation spaces are developed and analyzed in some detail and the stability of the resulting scheme is discussed. Section 3 is devoted to convergence experiments and investigates the performance of the method when applied to three different models. We conclude the paper by summarizing the various merits of the method.

2. Concepts and analysis

Since the spatial domain of the master equation is the set of non-negative integers and since no boundary conditions need to be imposed, a spectral method seems to be a favorable numerical tool. Spectral representations of stochastic processes have been used frequently in the context of *Stochastic differential equations* (SDEs) [25,30,31], but applying spectral methods directly to the master equation is a recent contribution. Most SDEs in applications are derived by adding noise to a deterministic description as a model of incomplete or inexact data and are expressed in observable variables. By contrast, the master equation directly governs the probability density function of the process and is a consequence of the Markov assumption only.

We now proceed by a discussion of the properties of the master equation. A construction of the approximation spaces then follows where "smoothness" of discrete functions will be defined and where care will be taken to ensure convergence in a relevant norm. We conclude by discussing the issue of stability.

2.1. The master equation

We shall consider the dynamics of a chemical system of D different species under R prescribed reactions. Let $p(x, t)$ be the probability distribution of the states $x \in \mathbf{Z}_+^D = \{0, 1, 2, \ldots\}^D$ at time t. That is, p simply describes the probability that a certain number of molecules is present at each time.

The reactions are specified as transitions between the states in $\mathbf{Z}_+^D$ according to the *reaction propensities* $w_r : \mathbf{Z}_+^D \to \mathbf{R}_+$. These define the transition probability per unit of time for moving from the state x to $x - \mathbb{N}_r$;

$$x \xrightarrow{w_r(x)} x - \mathbb{N}_r, \tag{2.1}$$

where $\mathbb{N}_r \in \mathbf{Z}^D$ is the transition step and is the rth column in the *stoichiometric matrix* $\mathbb{N}$.

The *master equation* [15,23] is then given by

$$\frac{\partial p(x,t)}{\partial t} = \sum_{\substack{r=1 \\ x+\mathbb{N}_r^- \geq 0}}^{R} w_r(x+\mathbb{N}_r)p(x+\mathbb{N}_r,t) - \sum_{\substack{r=1 \\ x-\mathbb{N}_r^+ \geq 0}}^{R} w_r(x)p(x,t) =: \mathcal{M}p, \tag{2.2}$$

where the transition steps are decomposed into positive and negative parts as $\mathbb{N}_r = \mathbb{N}_r^+ + \mathbb{N}_r^-$ and where the sums include *feasible* reactions only.

It is often preferable to work with the adjoint operator $\mathcal{M}^*$ [23, Ch. V.9]. If (p, q) is a pair of not necessarily normalized or positive functions defined over $\mathbf{Z}_+^D$, then provided both sides make sense (see [10] for a proof and an application to the moment problem),

$$\sum_{x\geq 0} q(x)\mathcal{M}p(x) = \sum_{x\geq 0}\sum_{r=1}^{R}[q(x-\mathbb{N}_r) - q(x)]w_r(x)p(x). \tag{2.3}$$

It follows that the adjoint operator is given by

$$\mathcal{M}^*q = \sum_{r=1}^{R} w_r(x)[q(x-\mathbb{N}_r) - q(x)]. \tag{2.4}$$

Note that both x and $\mathbb{N}_r$ in (2.3) and (2.4) are D-dimensional vectors and that the sum in (2.3) runs over $x \in \mathbf{Z}_+^D$. Other names for (2.2) and (2.4) include the *forward/backward Kolmogorov equations* [4, Ch. 2.3] and *forward/backward master equations* [17, Ch. 5.1.D].

Now let (λ, q) be an eigenpair of $\mathcal{M}^*$ normalized so that the largest value of q is positive and real. Then we see from (2.4) that $\Re\lambda \leq 0$ so that all eigenvalues of $\mathcal{M}$ share this property. Moreover, $q \equiv 1$ is an eigenvector corresponding to $\lambda = 0$, a fact that has the natural interpretation that the probability mass of any solution p is conserved by the master equation. We shall need the following related stability result.

Theorem 2.1. *Any solution to the master equation is non-increasing in the l^1-sequence norm. That is,*

$$\sum_{x\geq 0}|p(x,t)| \leq \sum_{x\geq 0}|p(x,0)| \tag{2.5}$$

for any $t \geq 0$.

Note that $p(x, 0)$ in Theorem 2.1 is an arbitrary l^1-measurable function defined on $\mathbf{Z}_+^D$, and not necessarily a probability. The statement follows from the fact that the semigroup corresponding to $\mathcal{M}$ is contractive (see [8, Ch. 1]), but in order to be self-contained we offer the following simple argument.

Proof. It is easier to prove this result by considering the adjoint equation under the dual norm of l^1, which is the l^∞-norm. This argument uses the equality $(q(\cdot,0), p(\cdot,t)) = (q(\cdot,t), p(\cdot,0))$ for the two solutions. From the relation $\partial q/\partial t = \mathcal{M}^*q$ and the definition (2.4) we see that the largest positive value of q cannot increase and that the most negative value cannot decrease (this observation is due to van Kampen, see the "remarkable exercise" in [23, Ch. V.9]). Consequently, $\|q\|_{l^\infty}$ cannot increase. In Section 2.5 a second and more direct proof of this result is obtained. □

Remark. It follows from Theorem 2.1 that for any $p \geq 1$, the l^p-norm of the solution stays bounded. However, for $p > 1$ it is frequently the case that the norm grows slowly in time.

We now cite the following important result which follows from more careful considerations of the structure of the master operator.

Theorem 2.2. *Let $p(x, 0)$ be an l^1-measurable discrete function defined on a bounded subset $S \subset \mathbf{Z}_+^D$ and assume that the master operator is restricted to functions on this domain. If $\mathcal{M}$ is neither decomposable nor a splitting (see below), then the master equation (2.2) admits a unique steady-state solution as $t \to \infty$. Moreover, if $p(x, 0)$ is a discrete probability density, then so is the steady-state solution.*

For a proof and an insightful discussion we refer the reader to [23, Ch. V.3]. A *decomposable* linear operator can be cast in the form (by relabeling the states)

$$\mathcal{M} = \begin{bmatrix} \mathcal{M}_{11} & 0 \\ 0 & \mathcal{M}_{22} \end{bmatrix}, \tag{2.6}$$

while a *splitting* operator can be written as

$$\mathcal{M} = \begin{bmatrix} \mathcal{M}_{11} & \mathcal{M}_{12} & 0 \\ 0 & \mathcal{M}_{22} & 0 \\ 0 & \mathcal{M}_{32} & \mathcal{M}_{33} \end{bmatrix}. \tag{2.7}$$

Exclusion of these cases essentially forces $\mathcal{M}$ to define a fully interacting system, one not allowed to consist of several isolated subsystems.

As indicated, Theorem 2.2 is only valid when the number of states is finite. Indeed, there are many examples of master equations on unbounded domains for which no steady-state solution exists (e.g. the standard Poisson process on the form $\emptyset \to X$ where X-molecules simply arrive indefinitely). For *chemical* master equations governing closed systems, however, each species must have sufficiently strong "sinks" to match the inflow from the "sources". We therefore expect reasoning based on assuming the existence of a steady-state solution to be valid for all physically realizable systems.

As a concluding model problem in one dimension we consider the linear birth–death process [2]

$$\left.\begin{array}{lcl} \emptyset & \xrightarrow{k} & X \\ X & \xrightarrow{\mu x} & \emptyset \end{array}\right\}, \tag{2.8}$$

where uppercase letters are used to denote molecule *names*, while lowercases are used for counting the number of molecules of the corresponding species. In (2.8), X-molecules are created at a constant rate and simultaneously destroyed at a rate proportional to the total number of molecules. The master equation for this system can be written in terms of the forward- and backward difference operator $\Delta q(x) = q(x+1) - q(x)$ and $\nabla q(x) = q(x) - q(x-1)$,

$$\frac{\partial p(x,t)}{\partial t} = \mathcal{M}p(x,t) = -k\bar{\nabla}p(x,t) + \mu\Delta[xp(x,t)], \tag{2.9}$$

where a bar over ∇ expresses the convention that $p(-1,t) = 0$. This problem can be solved completely if initial data is given in the form of a Poissonian distribution of expectation a_0,

$$p(x,0) = \frac{a_0^x}{x!}\mathrm{e}^{-a_0}. \tag{2.10}$$

In this case one easily verifies that the full dynamic solution is given by

$$p(x,t) = \frac{a(t)^x}{x!}\mathrm{e}^{-a(t)}, \tag{2.11}$$

where $a(t) = a_0\exp(-\mu t) + k/\mu\cdot(1-\exp(-\mu t))$. Independent of the initial data, p approaches a Poissonian distribution with expectation k/μ.

This example is part of the motivation for the unusual choice of basis functions suggested in the next section. *Charlier's* polynomials are orthogonal with respect to the Poisson process and appear promising as a means of representing solutions to the master equation. Approximation properties of this basis are investigated thoroughly in the next few sections.

2.2. Functions on $\mathbf{Z}_+$

In the following sections we shall study the approximation of functions defined over the set of non-negative integers. Suitable spaces of functions are introduced and investigated and a theory for approximation over these spaces is developed. The corresponding results for continuous approximation are fairly well understood but the discrete version seems to have been largely overlooked in the literature.

For clarity, we mention here that the theory will contain a certain parameter a and we make some efforts to obtain uniform results. This extra degree of freedom can be used as a means of improving the efficiency of the resulting scheme. To this end we shall only consider one-dimensional functions; the corresponding tensor basis is straightforward to develop.

For $p \in \{1, 2, \infty\}$ we will make use of the ordinary normed $l^p(\mathbf{Z}_+)$-spaces,

$$l^p(\mathbf{Z}_+) = \left\{q : \mathbf{Z}_+ \to \mathbf{R};\ \|q\|_{l^p(\mathbf{Z}_+)} < \infty\right\}, \tag{2.12}$$

$$\|q\|_{l^p(\mathbf{Z}_+)}^p \equiv \sum_{x\geq 0}|q(x)|^p, \tag{2.13}$$

where the usual sup-norm is to be understood when $p = \infty$. For $p = 2$ we additionally associate the discrete Euclidean inner product,

$$(p,q) \equiv \sum_{x\geq 0} p(x)q(x). \tag{2.14}$$

Define now the falling factorial power by $x^{\underline{m}} = x!/(x-m)! = \prod_{i=0}^{m-1}(x-i)$. For reasons that will be clear later on (see Lemma 2.4) we shall use the following hierarchy of discrete Sobolev-spaces:

$$h^m(\mathbf{Z}_+) = \left\{q : \mathbf{Z}_+ \to \mathbf{R};\ \sqrt{x^{\underline{k}}}\cdot q(x) \in l^2(\mathbf{Z}_+) \text{ for } 0 \leq k \leq m\right\} \tag{2.15}$$

with the corresponding norm

$$\|q\|^2_{h^m(\mathbf{Z}_+)} \equiv \sum_{k=0}^{m} a^{-k} \|\sqrt{x^{\underline{k}}} \cdot q(x)\|^2_{l^2(\mathbf{Z}_+)}, \tag{2.16}$$

and where the parameter $a \in \mathbf{R}_+$.

We also define an analogous set of *weighted* Sobolev-spaces with weight $w(x) = a^x/x! \cdot \mathrm{e}^{-a}$. The weighted inner product is

$$(p, q)_w \equiv \sum_{x\geq 0} p(x)q(x)w(x) \tag{2.17}$$

with generated norm. This yields the weighted space $l^2_w(\mathbf{Z}_+)$ and the definition of each weighted discrete Sobolev-space $h^m_w(\mathbf{Z}_+)$ follows as in (2.15) and (2.16). Since these spaces are less common in analysis, and in order to get some feeling for them, we consider the following example: set $p(x) = \sqrt{(x-2)!}$ with $p(0) = p(1) \equiv 0$ and let $a = 1$. Clearly, $p \in h^0_w$, and in fact $\|p\|^2_{h^0_w} = \exp(-1)$. However, $p \notin h^1_w$ by the divergence of the harmonic sum. As an easy generalization we note that $p(x)^2 = (x-m-2)!$ is in h^m_w but not in h^{m+1}_w.

We now further examine these spaces by proving the following two basic results concerning the forward- and backward difference operator.

Proposition 2.3. *The map $\Delta : h^{m+1}_w(\mathbf{Z}_+) \to h^m_w(\mathbf{Z}_+)$ is continuous uniformly w.r.t. to the parameter a.*

In fact, the following stronger result will be convenient later on and additionally provides us with some insight:

Lemma 2.4. *Define the norm*

$$\|q\|^2_{h^m_{w,\Delta}(\mathbf{Z}_+)} \equiv \sum_{k=0}^{m} \|\Delta^k q\|^2_{l^2_w(\mathbf{Z}_+)}. \tag{2.18}$$

Then the norms $\|\cdot\|_{h^m_{w,\Delta}(\mathbf{Z}_+)}$ and $\|\cdot\|_{h^m_w(\mathbf{Z}_+)}$ are uniformly equivalent. That is, there are positive constants C_1 and C_2 depending only on m such that

$$C_1\|q\|_{h^m_w(\mathbf{Z}_+)} \leq \|q\|_{h^m_{w,\Delta}(\mathbf{Z}_+)} \leq C_2\|q\|_{h^m_w(\mathbf{Z}_+)} \tag{2.19}$$

holds for any function $q \in h^m_w(\mathbf{Z}_+)$.

Proof. Denote the forward shift operator by $Eq(x) = q(x+1)$. We start by noting the useful relation

$$a^{-k}\|\sqrt{x^{\underline{k}}}q\|^2_{l^2_w} = \|E^k q\|^2_{l^2_w}.$$

Expanding $E^k = (I + \Delta)^k$ in binomial terms yields

$$\begin{aligned}\|E^k q\|^2_{l^2_w} &= \sum_{x\geq 0}\left(\sum_{j=0}^{k}\binom{k}{j}\Delta^j q(x)\right)^2 w(x)\\ &\leq \sum_{x\geq 0}\sum_{j=0}^{k}\binom{k}{j}^2\sum_{j=0}^{k}\left(\Delta^j q(x)\right)^2 w(x) \leq 4^k\|q\|^2_{h^k_{w,\Delta}}.\end{aligned}$$

Summing this for $k = 0 \ldots m$ gives the first bound with (say) $C_1^{-1} = 2^{m+1}$. The second bound with $C_2 = 2^{m+1}$ can be proved in exactly the same way, expanding $\Delta^k = (E - I)^k$ instead. □

The Sobolev-spaces generated by Δ are sometimes more convenient to work with and are perhaps more natural analogues to the standard continuous Sobolev-spaces. In Section 2.1 we defined the modified backward difference operator $\bar{\nabla}p(x) \equiv p(x-1) - p(x)$ with the exception of $\bar{\nabla}p(0) \equiv -p(0)$. Interestingly, there is no uniform (w.r.t. a) analogue of Proposition 2.3 for this operator. As a counter-example we note that the unit pulse at $x = 0$, i.e. $p(x) = 1$ if $x = 0$ and zero otherwise, yields $\|\bar{\nabla}p\|^2_{l^2_w} = (1+a)\exp(-a)$ whereas $\|p\|^2_{h^m_w} = \exp(-a)$ for *any* m. Thus, any bound on $\bar{\nabla}$ (or ∇) must be non-uniform with respect to a. In spite of this we prove the following partial result in this direction which will be helpful in order to bound a certain Sturm–Liouville operator to be introduced shortly.

Proposition 2.5. *The map $F : h^{m+2}_w(\mathbf{Z}_+) \to h^m_w(\mathbf{Z}_+)$ defined by $F(q) = x/a \cdot \nabla q$ is continuous. Furthermore, if $a \geq 1$, then the continuity is uniform with respect to this parameter.*

Proof. Split the operator according to $F(q) = x/a \cdot q - x/a \cdot E^{-1}q$, where E^{-1} is the backward shift operator (note that the convention $q(-1) = 0$ is not needed here). By definition the former map is continuous between h^{m+2}_w and h^m_w, although not necessarily uniformly so. Under the assumption $a \geq 1$, we expand for some constants A_k and B_k,

$x^2 = (x-k)(x-k-1) + A_k(x-k) + B_k$. Then

$$a^{-k}\|\sqrt{x^{\underline{k}}}\,x/a\cdot q\|_{l^2_w}^2 = a^{-(k+2)}\|\sqrt{x^{\underline{k+2}} + A_k x^{\underline{k+1}} + B_k x^{\underline{k}}}\cdot q\|_{l^2_w}^2,$$

and the bound is uniform with respect to a. As for the operator $x/a\cdot E^{-1}$, we proceed similarly for $k\geq 1$,

$$\begin{aligned} a^{-k}\|\sqrt{x^{\underline{k}}}\,x/a\cdot E^{-1}q\|_{l^2_w}^2 &= a^{-(k+2)}\sum_{x\geq 0}(x+1)^{\underline{k}}(x+1)^2 q(x)^2 w(x+1)\\ &= a^{-(k+1)}\sum_{x\geq 0}\left(x^{\underline{k+1}} + A_k x^{\underline{k}} + B_k x^{\underline{k-1}}\right) q(x)^2 w(x). \end{aligned}$$

A similar strategy for $k=0$ establishes that in fact, $x/a\cdot E^{-1} : h^{m+1}_w(\mathbf{Z}_+)\to h^m_w(\mathbf{Z}_+)$ is continuous (and uniformly so if $a\geq 1$). □

In contrast to Propositions 2.3 and 2.5, we note that by expanding $\Delta^m = (E-I)^m$ into binomials and using the triangle inequality we get $\|\Delta^m p\|_{l^2}\leq 2^m\|p\|_{l^2}$. Incidentally, this explains why the *unweighted* Sobolev-spaces h^m cannot be generated by Δ. A slight generalization of this fact will later be useful to bound the regularity of general master operators:

Proposition 2.6. *The maps $\bar\nabla : h^m(\mathbf{Z}_+)\to h^m(\mathbf{Z}_+)$ and $\Delta : h^m(\mathbf{Z}_+)\to h^m(\mathbf{Z}_+)$ are continuous.*

Proof. We only consider the result for Δ since the proof of the other case is similar. By writing $\Delta = E-I$ it suffices to prove the boundedness of the forward shift operator E. Define $\bar q(x) = q(x)$ except for $\bar q(0)\equiv 0$ and note that by inspection,

$$\|\sqrt{x^{\underline{k}}}Eq\|_{l^2} = \|\sqrt{(x-1)^{\underline{k}}}\bar q\|_{l^2} = \|\sqrt{x^{\underline{k}} - kx^{\underline{k-1}}}\bar q\|_{l^2}\leq \|\sqrt{x^{\underline{k}}}q\|_{l^2},$$

with the exception of the trivial case $k=0$. □

2.3. Charlier approximation

We now let $C^a_n(x)$ denote the *normalized* nth degree Charlier polynomial [24] with parameter $a>0$. These polynomials form an orthonormal set of functions with respect to the l^2_w-product; $(C^a_n, C^a_m)_w = \delta_{nm}$. We write X_N for the span of the (Charlier-) polynomials of degree $\leq N$ and define π_N as the orthogonal projection onto X_N associated with $(\cdot,\cdot)_w$.

The normalized Charlier polynomials satisfy the recurrence

$$\begin{aligned} C^a_0(x) &\equiv 1,\\ C^a_1(x) &\equiv \frac{a-x}{\sqrt{a}},\\ C^a_{n+1}(x) &= \frac{n+a-x}{\sqrt{a(n+1)}}C^a_n(x) - \sqrt{\frac{n}{n+1}}C^a_{n-1}(x). \end{aligned} \tag{2.20}$$

There is a also a Charlier difference equation,

$$SC^a_n(x) := -w^{-1}(x)\nabla\left[w(x)\Delta C^a_n(x)\right] = \frac{n}{a}C^a_n(x), \tag{2.21}$$

where the Sturm–Liouville operator S can be expanded as

$$Sp = \frac{x}{a}\nabla p - \Delta p. \tag{2.22}$$

Interestingly, this is in fact the dual to the birth–death operator (2.9). Charlier's polynomials are compatible with the forward difference operator in the sense that

$$\Delta C^a_n(x) = -\sqrt{n/a}\cdot C^a_{n-1}(x). \tag{2.23}$$

As is well-known in Sturm–Liouville theory, the approximation properties of orthogonal functions depend crucially on the regularity of the corresponding Sturm–Liouville operator. This is the motivation for our interest in Propositions 2.3 and 2.5 since they immediately yield (cf. the expanded form (2.22) of the operator S),

Lemma 2.7. *The operator $S : h^{m+2}_w(\mathbf{Z}_+)\to h^m_w(\mathbf{Z}_+)$ is continuous and thus bounded. If $a\geq 1$ is assumed, then the continuity is uniform with respect to this parameter.*

Recall now the summation by parts formula in the following form:

$$\sum_{x=0}^{N} p(x)\Delta q(x) = p(N)q(N+1) - p(-1)q(0) - \sum_{x=0}^{N}\nabla p(x)q(x), \tag{2.24}$$

where usually we will have that both boundary terms vanish. The following lemma relates the coefficients of an orthogonal expansion in terms of Charlier polynomials with the Sturm–Liouville operator S.

Lemma 2.8. *Let $p \in h^m_w(\mathbf{Z}_+)$. Then*

$$(m \text{ even}) \quad (p, C^a_n)_w = (a/n)^{m/2} \left(S^{m/2}p, C^a_n\right)_w, \tag{2.25}$$

$$(m \text{ odd}) \quad (p, C^a_n)_w = -(a/n)^{m/2} \left(\Delta S^{(m-1)/2}p, C^a_{n-1}\right)_w. \tag{2.26}$$

Proof. In view of (2.21) we get

$$(p, C^a_n)_w = -\frac{a}{n}\left(p, \nabla\left[w\Delta C^a_n\right]\right).$$

Summation by parts then yields in turn

$$= \frac{a}{n}\left(\Delta p, w\Delta C^a_n\right) = -\frac{a}{n}\left(\nabla\left[w\Delta p\right], C^a_n\right) = \frac{a}{n}\left(Sp, C^a_n\right)_w.$$

If m is even, repeating this procedure a total of $m/2$ times concludes the proof of (2.25). For the odd case we continue from

$$\begin{aligned}(a/n)^{(m-1)/2}\left(S^{(m-1)/2}p, C^a_n\right)_w &= -(a/n)^{(m+1)/2}\left(S^{(m-1)/2}p, \nabla\left[w\Delta C^a_n\right]\right)\\ &= (a/n)^{(m+1)/2}\left(\Delta S^{(m-1)/2}p, \Delta C^a_n\right)_w.\end{aligned}$$

Using (2.23) now produces (2.26). □

Theorem 2.9. *For any nonnegative integer m, there exists a positive constant C depending only on m and a such that, for any function $p \in h^m_w(\mathbf{Z}_+)$, the following estimate holds*

$$\|\pi_{N-1}p - p\|_{l^2_w(\mathbf{Z}_+)} \le C(a/N)^{m/2}\|p\|_{h^m_w(\mathbf{Z}_+)}. \tag{2.27}$$

If in addition, $a \ge 1$ is assumed, then C depends only on m.

Proof. Expanding any function $p \in l^2_w$ in terms of Charlier polynomials, we readily get

$$\|\pi_{N-1}p - p\|^2_{l^2_w} = \sum_{n\ge N}\bar{p}^2_n,$$

where, provided m is an even integer, we have by Lemma 2.8 that

$$\bar{p}_n = (p, C^a_n)_w = (a/n)^{m/2}\left(S^{m/2}p, C^a_n\right)_w.$$

Hence,

$$\|\pi_{N-1}p - p\|^2_{l^2_w} \le (a/N)^m \sum_{n\ge N}\left(S^{m/2}p, C^a_n\right)^2_w \le (a/N)^m\|S^{m/2}p\|^2_{l^2_w}.$$

Lemma 2.7 thus concludes the even case. When m is an odd integer we proceed similarly, using instead the odd version of Lemma 2.8. □

Theorem 2.9 is reminiscent of results for continuous approximations. See for example Theorem 12.1 in [3] (p. 289) for approximating continuous functions over $\mathbf{R}_+$ by Laguerre polynomials. Worth noting with the continuous theory is the technical need for an additional hierarchy of Sobolev-spaces (cf. equation 12.6 in [3], p. 288). This can be avoided completely in the present case thanks to Lemma 2.4.

We are now in the position to consider approximation in stronger norms. The following lemma makes this possible although we would like to point out that the given bound is very weak and can easily be improved upon. It seems, however, that such improvements only complicate what follows.

Lemma 2.10. *For a constant C depending only on m,*

$$\|C^a_n\|_{h^m_w(\mathbf{Z}_+)} \le C\max(1, n/a)^{m/2}. \tag{2.28}$$

Proof. It is easier to prove this using the norm $\|\cdot\|_{h^m_{w,\Delta}(\mathbf{Z}_+)}$. From (2.23) we immediately get

$$\|\Delta^k C^a_n\|^2_{l^2_w} = \frac{n^{\underline{k}}}{a^k} \le \frac{n^k}{a^k}.$$

Summation yields $\|C^a_n\|_{h^m_{w,\Delta}} \le \sqrt{m}\max(1, n/a)^{m/2}$ and the bound follows. □

This "smoothness" of the basis polynomials yields the following generalization of Theorem 2.9:

Theorem 2.11. *For any nonnegative integers k and m, $k \leq m$, there exists a positive constant C depending only on m and a such that, for any function $p \in h^m_w(\mathbf{Z}_+)$, the following estimate holds*

$$\|\pi_{N-1}p - p\|_{h^k_w(\mathbf{Z}_+)} \leq C(a/N)^{m/2} \max(1, N/a)^{k/2} \|p\|_{h^m_w(\mathbf{Z}_+)}. \tag{2.29}$$

Again, C depends only on m if $a \geq 1$ is assumed.

Proof. Again it is convenient to construct the proof in the uniformly equivalent norm $\|\cdot\|_{h^m_{w,\Delta}(\mathbf{Z}_+)}$. The case $k = 0$ corresponds to Theorem 2.9 and we proceed by induction, assuming that (2.29) holds for some k. Split the error according to

$$\begin{aligned}\|\pi_{N-1}p - p\|_{h^{k+1}_{w,\Delta}} &\leq \|\pi_{N-1}p - p\|_{l^2_w} + \|\pi_{N-1}\Delta p - \Delta p\|_{h^k_{w,\Delta}} + \|\Delta\pi_{N-1}p - \pi_{N-1}\Delta p\|_{h^k_{w,\Delta}} \\ &\leq C_1(a/N)^{m/2}\|p\|_{h^m_{w,\Delta}} + C_2(a/N)^{(m-1)/2-k/2}\|\Delta p\|_{h^{m-1}_{w,\Delta}} + \|\Delta\pi_{N-1}p - \pi_{N-1}\Delta p\|_{h^k_{w,\Delta}},\end{aligned} \tag{2.30}$$

where Theorem 2.9 and the induction hypothesis have been used. Evidently, in this norm we have that $\|\Delta p\|_{h^{m-1}_{w,\Delta}} \leq \|p\|_{h^m_{w,\Delta}}$, and so we focus on the last term. Writing as before

$$p = \sum_{n\geq 0} \bar{p}_n C^a_n,$$

we readily obtain

$$\Delta\pi_{N-1}p = \sum_{n=0}^{N-1} \bar{p}_n \Delta C^a_n \quad \text{and} \quad \pi_{N-1}\Delta p = \sum_{n=0}^{N} \bar{p}_n \Delta C^a_n.$$

The last term in (2.30) is therefore

$$\|\bar{p}_N \Delta C^a_N\|_{h^k_{w,\Delta}} \leq |\bar{p}_N| \|C^a_N\|_{h^{k+1}_{w,\Delta}} \leq C_3 |(p, C^a_N)_w| \max(1, N/a)^{(k+1)/2},$$

where Lemma 2.10 was used. By Lemma 2.8, Cauchy–Schwarz's inequality, and Lemma 2.7 this becomes

$$\leq C_4(a/N)^{m/2} \max(1, N/a)^{(k+1)/2} \|p\|_{h^m_{w,\Delta}},$$

finishing the induction step. □

In other words, the cost for measuring the error in the stronger norm $\|\cdot\|_{h^k_w(\mathbf{Z}_+)}$ is determined by the regularity of the basis, a situation that again is encountered in many continuous settings (for the corresponding result for Laguerre polynomials, see Theorem 12.3 in [3], p. 291).

We now take a look at approximation in the unweighted Sobolev-spaces $h^m(\mathbf{Z}_+)$. Define *Charlier's functions* by $\hat{C}^a_n(x) := C^a_n(x) \cdot w(x)^{1/2}$ along with the space $\hat{X}_N = \{p(x) = q(x) \cdot w(x)^{1/2};\ q \in X_N\}$. Evidently, these functions are orthonormal under the usual l^2-product $(\cdot, \cdot)$ and we use $\hat{\pi}_N$ to denote the corresponding orthogonal projection on $\hat{X}_N$. The relation

$$\hat{\pi}_N p = w(x)^{1/2} \pi_N \left(p(x) \cdot w(x)^{-1/2}\right) \tag{2.31}$$

is immediate and we make the crucial observation that the map $p \to w^{1/2}p$ is an *isomorphism* between h^m_w and h^m. This implies the following result.

Corollary 2.12. *For any nonnegative integers k and m, $k \leq m$, there exists a positive constant C depending only on m and a (or only on m provided $a \geq 1$ is given) such that, for any function $p \in h^m(\mathbf{Z}_+)$, the following estimate holds*

$$\|\hat{\pi}_{N-1}p - p\|_{h^k(\mathbf{Z}_+)} \leq C(a/N)^{m/2} \max(1, N/a)^{k/2} \|p\|_{h^m(\mathbf{Z}_+)}. \tag{2.32}$$

This result implies that an efficient spectral representation exists and that expanding a smooth solution in the space $\hat{X}_N$ drastically may reduce the number of degrees of freedom when compared to e.g. a direct representation. Corollary 2.12 is again related to similar results for continuous approximation; see for example [27].

There are several reasons for preferring to seek approximations to solutions of the master equation in the space $\hat{X}_N$ rather than in X_N. First, any Galerkin formulation of the master equation in the inner product $(\cdot, \cdot)_w$ will at best lead to convergence in the corresponding norm $\|\cdot\|_{l^2_w}$. In contrast, a convergent Galerkin formulation in the l^2-product will of course imply the existence of error estimates in the much stronger l^2-norm. Second, solutions in X_N are not probability distributions and statistical functionals of interest, such as the mean and variance, can therefore not be computed.

2.4. Conservation of probability

Unfortunately, the projection $\hat{\pi}_N$ is not sufficiently conservative for our present purposes. The reason is that it does not preserve the probability mass; in general we have that $(1, \hat{\pi}_N p) \neq (1, p) = 1$. For the projection to be conservative we need to somehow enforce the preservation of total probability. We therefore consider the projection $\hat{\pi}^0_N p = p_N$ which for some

Lagrange multiplier λ satisfies

$$\left.\begin{array}{l}(q, p_N - p) + \lambda(f(q), 1) = 0\\ (1, p_N - p) = 0\end{array}\right\} \quad \text{for } \forall q \in \hat{X}_N, \tag{2.33}$$

where f is a suitable nonzero linear function to be determined. To analyze this projection, we first note that, regardless of the norm,

$$\|\hat{\pi}_N^0 p - p\| \le \|\hat{\pi}_N^0 p - \hat{\pi}_N p\| + \|\hat{\pi}_N p - p\| \tag{2.34}$$

and that as consequences of (2.33),

$$\hat{\pi}_N^0 p - \hat{\pi}_N p = \sum_{n=0}^{N} (\tilde{p}_n - \bar{p}_n)\hat{C}_n^a = -\lambda \sum_{n=0}^{N} (f(\hat{C}_n^a), 1)\hat{C}_n^a, \tag{2.35}$$

$$(1, \hat{\pi}_N p - p) = -(1, \hat{\pi}_N^0 p - \hat{\pi}_N p) = \lambda \sum_{n=0}^{N} (f(\hat{C}_n^a), 1)(\hat{C}_n^a, 1), \tag{2.36}$$

where $\tilde{p}_n$ and $\bar{p}_n$ are the coefficients produced by $\hat{\pi}_N^0$ and $\hat{\pi}_N$, respectively. The l^2-deviation between $\hat{\pi}_N^0$ and the orthogonal projection $\hat{\pi}_N$ is therefore generally given by

$$\|\hat{\pi}_N^0 p - \hat{\pi}_N p\|_{l^2}^2 = \lambda^2 \sum_{n=0}^{N} (f(\hat{C}_n^a), 1)^2. \tag{2.37}$$

For the traditional Lagrangian choice $f(q) = q$, (2.35) and (2.36) can be combined into

$$\|\hat{\pi}_N^0 p - \hat{\pi}_N p\|_{l^2}^2 = \frac{(1, \hat{\pi}_N p - p)^2}{\sum_{n=0}^{N} (\hat{C}_n^a, 1)^2}. \tag{2.38}$$

However, this projection is a somewhat inconvenient choice in a Galerkin formulation of the time-dependent master equation. Modifying all frequencies $\hat{C}_n^a$ is more complicated to implement and does not yield a much smaller error than modifying the lowest frequency $\hat{C}_0^a$ alone. This corresponds to the choice $f(q) = \hat{\pi}_0 q$ and satisfies (from (2.35) and (2.36))

$$\|\hat{\pi}_N^0 p - \hat{\pi}_N p\|_{l^2}^2 = \frac{(1, \hat{\pi}_N p - p)^2}{(\hat{C}_0^a, 1)^2}. \tag{2.39}$$

Although the error (2.39) is slightly larger than (2.38), it is found experimentally that $\hat{C}_0^a$ carries more mass than the rest of the modes. Consequently, the sum in the denominator of (2.38) is replaced by the largest term so that in practice the difference is not critical. Remarkably then, using the "tau-method" [22], or what amounts to the same thing, making the choice $f(q) = (\hat{C}_N^a, q)\hat{C}_N^a$, cannot be recommended. The corresponding deviation has the same appearance as (2.39), but with the denominator replaced by $(\hat{C}_N^a, 1)^2$. Since the mass of $\hat{C}_N^a$ is smaller than that of $\hat{C}_0^a$, this method performs worse than the suggested projection.

Another feature of the choice $f(q) = \hat{\pi}_0 q$ is that a reasonably sharp error estimate in the l^1-norm is easily obtained. By inspection $\hat{C}_0^a$ is positive everywhere so that $(1, \hat{C}_0^a) = \|\hat{C}_0^a\|_{l^1}$. Hence from (2.34)–(2.36),

$$\|\hat{\pi}_N^0 p - p\|_{l^1} \le |(1, \hat{\pi}_N p - p)| + \|\hat{\pi}_N p - p\|_{l^1} \le 2\|\hat{\pi}_N p - p\|_{l^1}. \tag{2.40}$$

2.5. Stability

The Galerkin approximation to (2.2) that we shall now analyze reads as follows (compare (2.33)): find $p_N \in \hat{X}_N$ such that

$$\left.\begin{array}{l}(q, \partial p_N/\partial t) + \lambda(\hat{\pi}_0 q, 1) = (q, \mathcal{M} p_N)\\ (1, \partial p_N/\partial t) = 0\end{array}\right\} \quad \text{for } \forall q \in \hat{X}_N. \tag{2.41}$$

Since $\mathcal{M}$ generally is unbounded, indefinite and non-symmetric with non-orthogonal eigenvectors, we cannot possibly hope to capture any convergence properties of (2.41) by adhering to standard energy estimates. The partial results we present in this section are instead based on observations due to van Kampen [23, Ch. V] and Theorem 2.1. We will attempt to make it reasonable to believe that (2.41) is stable in the l^1-norm so that convergence follows from the Lax–Richtmyer equivalence theorem.

We first write (2.41) in the equivalent form

$$\frac{\partial p_N}{\partial t} = \hat{\pi}_N^0 \mathcal{M} p_N, \tag{2.42}$$

where the representation

$$p_N(x,t) = \sum_{n=0}^{N} c_n(t)\hat{C}_n^a(x) \tag{2.43}$$

is implicitly understood.

For a not necessarily positive or normalized p_N, let $U_N(t)$ denote the sum of the positive elements,

$$U_N(t) \equiv \sum_{x\geq 0} p_N(x,t)^+ = \sum_{x\in\xi^+(t)} p_N(x,t), \tag{2.44}$$

and similarly for $V_N(t)$, the sum of the negative elements. A crucial property of the master operator is that it preserves the probability mass and, since $\hat{\pi}_N^0$ is used rather than $\hat{\pi}_N$, this property holds true for $\hat{\pi}_N^0\mathcal{M}$ as well. Thus,

$$U_N(t) + V_N(t) = \text{constant}. \tag{2.45}$$

The time derivative of $U_N(t)$ exists between the events when $p_N(x,t)$ changes sign for some x. In such intervals we have that

$$\begin{aligned} U_N'(t) &= \sum_{x\in\xi^+(t)} \hat{\pi}_N^0\mathcal{M}p_N = \sum_{x\in\xi^+(t)} \mathcal{M}p_N + \sum_{x\in\xi^+(t)} \left[\hat{\pi}_N^0\mathcal{M}p_N - \mathcal{M}p_N\right] \\ &=: A_N(t) + B_N(t), \end{aligned} \tag{2.46}$$

where from (2.2),

$$A_N(t) = \sum_{x\in\xi^+(t)} \sum_{r=1}^{R} \left[x \geq -\mathbb{N}_r^-\right] w_r(x+\mathbb{N}_r)p_N(x+\mathbb{N}_r,t) - w_r(x)p_N(x,t), \tag{2.47}$$

and where the notation $[f]$, with f a logical expression, is used according to $[f] \equiv 1$ if f and $\equiv 0$ otherwise.

We always have that $A_N(t) \leq 0$. To see why, note that the sum over $x \in \xi^+(t)$ in (2.47) is built up by *some* of the positive elements of $w_r p_N$ minus *all* of its positive elements.

In the analytical case when B_N vanishes this constitutes a second proof of Theorem 2.1, since $\|p\|_{l^1} = U - V$ and, by (2.45), $-V' = U'$. When for some x, $p(x,t)$ changes sign, then although the derivative does not exist, the l^1-norm still depends continuously on time and consequently it cannot increase.

For the numerical investigation to follow, we see that

$$\frac{\mathrm{d}}{\mathrm{d}t}\|p_N\|_{l^1} = 2A_N(t) + 2B_N(t), \tag{2.48}$$

except for those points in time where a change of sign occurs. We would like to claim that *for N sufficiently large,* $|A_N| \geq |B_N|$, *so that the* l^1*-norm of the numerical solution does not increase*.

Although perhaps not a general proof of the claim, the motivation is that $\hat{\pi}_N^0\mathcal{M}p_N$ approaches $\mathcal{M}p_N$ when N grows so that in principle B_N can be made arbitrarily small. We are then clearly interested in the cases when A_N is zero, since this could induce a growth of the l^1-norm. Three cases are trivial: either one of $\xi^+, \xi^- := \{x;\ p_N(x,t) < 0\}$ or $\xi^0 := \{x;\ p_N(x,t) = 0\}$ is identical to $\mathbf{Z}_+$ so that p_N is either positive, negative or zero. In all these cases B_N vanishes as well since the mass-preserving projection is used — hence the l^1-norm must stay constant. Two cases force a closer examination of the master operator: (i) ξ^0 is empty while ξ^+ and ξ^- are not, and, (ii) all the sets ξ^+, ξ^- and ξ^0 are non-empty.

Case (i) Relabel the states so that p_N is divided into a positive and a negative part. Then we have

$$\mathcal{M} = \begin{bmatrix} \mathcal{M}_{++} & 0 \\ 0 & \mathcal{M}_{--} \end{bmatrix}, \tag{2.49}$$

for otherwise A_N cannot be zero (the zero in the lower left corner stems from using $-V' = U'$ and writing down the analogues of (2.46) and (2.47) for V_N). Hence $\mathcal{M}$ is *decomposable* (see (2.6)) and can be excluded from the present context since it does not describe a fully interacting chemical system but rather contains two isolated systems. Again, these considerations are formally only valid for a finite number of states (see the discussion in the end of Section 2.1).

Case (ii) Split the states of p_N into a positive, an all-zero, and a negative part, respectively. Accordingly,

$$\mathcal{M} = \begin{bmatrix} \mathcal{M}_{++} & \mathcal{M}_{+0} & 0 \\ 0 & \mathcal{M}_{00} & 0 \\ 0 & \mathcal{M}_{-0} & \mathcal{M}_{--} \end{bmatrix}. \tag{2.50}$$

That is, $\mathcal{M}$ is a *splitting* (see (2.7)) and can be excluded from the discussion for the same reason as above.

In conclusion then, A_N can only be zero when B_N is simultaneously zero (indicating a constant l^1-norm of p_N). If this is not the case, then we have that $A_N < 0$ and B_N tends to zero with increasing N. To look at the possible dependence of the

magnitudes of these two terms, we write out the derivative of the l^1-norm explicitly (by expanding $U'_N - V'_N$ as in (2.46)),

$$\frac{\mathrm{d}}{\mathrm{d}t}\|p_N\|_{l^1} = \underbrace{\sum_{x\geq 0} \operatorname{sgn} p_N\, \mathcal{M}p_N}_{2A_N} + \underbrace{\sum_{x\geq 0} \operatorname{sgn} p_N \left[\hat{\pi}^0_N \mathcal{M}p_N - \mathcal{M}p_N\right]}_{2B_N}, \tag{2.51}$$

where sgn q is zero for $q = 0$. It seems reasonable to believe that there are estimates of the form $2|A_N| \geq \kappa(\mathcal{M})\|\mathcal{M}p_N\|_{l^1}$ and $2|B_N| \leq C_N\|\mathcal{M}p_N\|_{l^1}$, where $\kappa(\mathcal{M})$ is a constant depending on the structure of the master operator and where C_N tends to zero when N increases. Under these assumptions, the derivative of the l^1-norm is $\leq (C_N - \kappa(\mathcal{M}))\|\mathcal{M}p_N\|_{l^1}$ which for N sufficiently large is less than or equal to zero. This argument does not prove l^1-stability unless the indicated estimates are first proved but it does, however, shed some light on the expected stability properties of the Galerkin scheme (2.41). Also, the crucial argument in this discussion is the relation (2.45) which indicates why a mass-preserving projection is a favorable choice.

3. Numerical experiments

In this section the feasibility of the proposed method is demonstrated by numerically solving three different models. Two systems with known solutions are used to illustrate the application of the theory and the numerical convergence. The third model is two-dimensional without known solutions and models the reaction between two species.

The method was implemented using *Gauss–Charlier quadratures* [11];

$$\sum_{x\geq 0} f(x)\frac{a^x}{x!}\mathrm{e}^{-a} = \sum_{j=1}^{n} f(x_j)w_j + R_n, \tag{3.1}$$

$$R_n = a^n n!\frac{f^{(2n)}(\xi)}{(2n)!}, \quad \xi \in (0,\infty). \tag{3.2}$$

The x_j's are the roots of $C^a_n(x)$ and the weights can be computed according to the formula

$$w_j \equiv -\frac{(an)^{-1/2}}{C^a_{n-1}(x_j)\cdot \mathrm{d}/\mathrm{d}x\, C^a_n(x_j)}. \tag{3.3}$$

In all experiments, the mass-preserving projection was used and the `Matlab` ODE-solver `ode15s` was employed for the evolution in time of the Galerkin spectral coefficients.

3.1. Application of the theory in one dimension

In order to highlight the application of the theory developed in Sections 2.2 and 2.3 we first consider two one-dimensional models with known solutions. The first is the linear birth–death problem (2.8) with time-dependent solution given by (2.11) and the second is the following set of reactions characterizing spontaneous bimolecular decay:

$$\left.\begin{array}{rcl} \emptyset & \xrightarrow{k} & X \\ X+X & \xrightarrow{\nu x(x-1)} & \emptyset \end{array}\right\}. \tag{3.4}$$

The master equation for this model can be written compactly as

$$\frac{\partial p(x,t)}{\partial t} = -k\bar{\nabla}p(x,t) + \nu\left[\Delta^2 + 2\Delta\right][x(x-1)p(x,t)], \tag{3.5}$$

where again the bar over the backward difference operator ∇ expresses the convention that $p(-1,t) = 0$. The full time-dependent solution to (3.5) is not known but the steady-state solution can be expressed in terms of the modified Bessel function I_n [23, X.2]. Let $a = \sqrt{k/\nu}$ and define $C = \sqrt{2}I_1(2\sqrt{2}a)$. Then

$$p(x,\infty) = C^{-1}\frac{a^x}{x!}I_{x-1}(2a) \tag{3.6}$$

is the exact stationary solution to (3.5). The expectation value m and the variance v can now be determined from standard asymptotics [1]:

$$m(\infty) \sim \sqrt{\frac{k}{2\nu}} + \frac{1}{8} + \mathcal{O}\left(a^{-1}\right), \tag{3.7}$$

$$v(\infty) \sim \frac{3}{4}\sqrt{\frac{k}{2\nu}} + \frac{1}{16} + \mathcal{O}\left(a^{-1}\right). \tag{3.8}$$

In order to apply the theory of Section 2.2 we first note that the master operator in (2.9) is bounded when regarded as an operator $h^{m+2} \to h^m$. We already know from Proposition 2.6 how the backward- and forward differences behave and it is

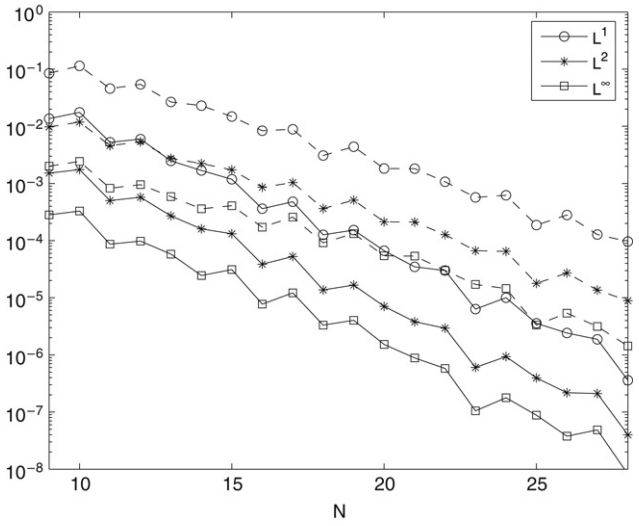

Fig. 3.1. Errors of the scheme applied to (2.8) (solid, time-average error) and (3.4) (dashed, error at steady-state) in different norms.

clear by definition that $p \to xp$ is bounded as a map $h^{m+2} \to h^m$. The situation is similar in (3.5). Here the quadratic factor $x(x-1)$ decreases the regularity by four degrees so that the master operator (3.5) is bounded as a map $h^{m+4} \to h^m$.

The analysis thus conducted is very general. Firstly, note that the reaction steps are always finite so that the master equation can always be written as a linear combination of terms that are finite compositions of difference operators. Secondly, *atomic* propensities that follow from combinatorial arguments are always low order polynomials, easily bounded as shown above. More general non-atomic propensities are also common. For instance, when quasi steady-state approximations are used for the derivation of a reduced model, the result is a set of rational functions involving roots of polynomials. These expressions can generally be majorized by low order polynomials and the resulting master operator can again be bounded as outlined above.

The parts of the analysis that we have to leave in a more incomplete state are the stability of the scheme and the regularity of the solution. The stability was discussed in Section 2.5 and we will simply assume that the scheme is stable. Moreover, regularity estimates are trivially available here since we do know the exact solutions. In the general case, however, conditions on the master operator in D dimensions for a certain degree of regularity of the solution seem very difficult to specify.

The numerical experiments were conducted as follows. We let the parameters be defined by $[k, \mu] = [1, 10^{-2}]$ in (2.8) and $[k, \nu] = [1, 5 \times 10^{-5}]$ in (3.4). This makes the expectation value in steady-state to be $m = 100$ very closely for both models. For (2.8), we start at $t = 0$ with a Poisson distribution with mean 75 and evolve the system until time $t = 10^4$. By comparing the solution thus obtained to the exact solution, the time-averaged error was determined. For the non-linear problem (3.4), since the solution is known at steady-state only, a Poisson distribution of mean 100 was used as the initial data. Again the system was evolved until $t = 10^4$ and the solution at this point was considered to be in steady-state. For both problems, the parameter a was taken to be 100.

In Fig. 3.1, errors in various measures are shown and it is clear that the convergence is exponential in the order N of the scheme. The slightly larger error for the non-linear problem (3.5) can be explained by the lesser regularity of both the solution and the corresponding master operator.

3.2. A two-dimensional example

As one of the simplest possible examples involving a reaction between two species X and Y we consider the following model:

$$\left.\begin{array}{rcl} \emptyset & \xrightarrow{k} & X \\ X & \xrightarrow{\mu x} & \emptyset \\ \emptyset & \xrightarrow{k} & Y \\ Y & \xrightarrow{\mu y} & \emptyset \\ X+Y & \xrightarrow{\nu xy} & \emptyset \end{array}\right\}. \tag{3.9}$$

The parameters are $[k, \mu, \nu] = [1, 2 \times 10^{-3}, 10^{-3}]$ so that the solution in steady-state is a symmetric distribution of mean about 32 in both variables with a strong negative correlation. Since an increase in one of the variables implies a decrease in the other by means of the bimolecular reaction, the negative correlation is expected from first principles.

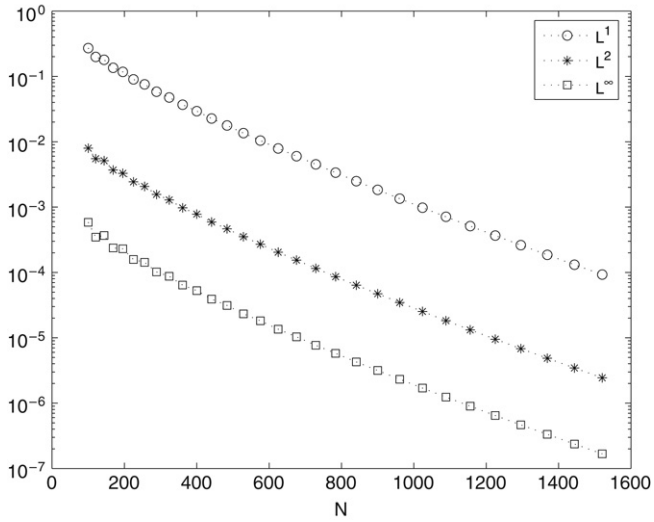

Fig. 3.2. Errors of the scheme applied to (3.9) in different norms versus the total number of degrees of freedom.

The master operator for this problem can be written in the notation used previously,

$$\mathcal{M} = -k[\bar{\nabla}_x + \bar{\nabla}_y] + \mu[\Delta_x x + \Delta_y y] + \nu[\Delta_x + \Delta_x\Delta_y + \Delta_y]xy. \tag{3.10}$$

For this problem we started the simulation with a two-dimensional Poisson distribution of mean 20 in both coordinates and evolved the system until time $t = 4000$. The parameter a was taken to be 20 for this case and a very high order reference solution was constructed using a more stringent error tolerance for the time-stepper. In Fig. 3.2 the time-averaged error is displayed in different norms. The exponential convergence is clearly visible.

In order to fully conclude the feasibility of the proposed spectral method we would have to construct in detail a tensorized generalization of the spaces developed in Section 2.2. This is beyond the scope here but we offer a few hints as to how to this can be achieved. Notice first that the argument in the proof of Proposition 2.6 holds with $\sqrt{x^{\underline{k}}}E$ replaced with (say) $\sqrt{x^{\underline{k_1}}y^{\underline{k_2}}}E_xE_y$. One therefore obtains that for instance the mapping $p \to \nu\Delta_x\Delta_y xy\,p$ as found among the terms in (3.10) can be bounded as a map between Sobolev-spaces of functions over $\mathbf{Z}_+^2$. In this way, all master operators formed from atomic propensities can readily be bounded in a tensor product version of the spaces developed in Section 2.2.

In addition, it seems very likely that there are highly general conditions for when the regularity of the solution remains at least at the same degree as the initial data during any interval (finite or infinite) of integration. Under the assumption of stability we then expect exponential convergence of the proposed scheme.

4. Conclusions

The master equation is a stochastic description of general discrete dynamical systems and is equivalent to the Markov assumption. It applies in particular to chemical reactions where the system is described by counting the number of molecules of each kind. Stochastic descriptions are preferred for many chemical systems of interest; examples can be found inside living cells where the effects of stochasticity are important.

We have proposed a novel spectral method for the master equation based on Charlier functions. Key features include high accuracy even at a relatively low resolution per dimension and convergence in the full semi-infinite discrete state–space. Stability and convergence is illustrated through numerical experiments suggesting that the scheme is effective when the dimensionality of the problem is not too high. The curse of dimension is of course still an issue, but a spectral expansion is a more efficient representation than is the full solution.

After this work was completed, the author became aware of a similar method for polyreaction kinetics devised by Deuflhard and Wulkow [7,28,29]. Their original scheme uses the basis $\{\tilde{C}_n^a(x)\} \coloneqq \{C_n^a(x) \cdot w(x)\}$ and determines the coefficients by taking the l^2-product with $\{C_n^a(x)\}$ in a Petrov–Galerkin formulation. The setting is one-dimensional and the model problem treated is reminiscent of the birth–death problem (2.8). However, when applied to the master equation of dimensionality higher than one, their scheme does not seem to be stable with the method of lines discretization. A related recent work directly aimed at the master equation is found in [6].

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