Defect-based local error estimators for splitting methods, with application to Schrödinger equations, Part II. Higher-order methods for linear problems[1]

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Abstract

In this work, defect-based local error estimators for higher-order exponential operator splitting methods are constructed and analyzed in the context of time-dependent linear Schrödinger equations. The technically involved procedure is carried out in detail for a general three-stage third-order splitting method and then extended to the higher-order case. Asymptotical correctness of the a posteriori local error estimator is proven under natural commutator bounds for the involved operators, and along the way the known (non)stiff order conditions and a priori convergence bounds are recovered. The theoretical error estimates for higher-order splitting methods are confirmed by numerical examples for a test problem of Schrödinger type. Further numerical experiments for a test problem of parabolic type complement the investigations.

Keywords: Linear evolution equations, Time-dependent linear Schrödinger equations, Time integration, Higher-order exponential operator splitting methods, Defect correction, A priori local error estimates, A posteriori local error estimates.

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1. Introduction

Scope of applications. A variety of contributions confirm the favorable performance of exponential operator splitting methods [1, 2] for the time integration of evolutionary Schrödinger equations [3]. As a small selection of works providing numerical evidence and a profound theoretical error analysis for linear and nonlinear problems, we refer to [4, 5, 6, 7, 8] and references given therein. Moreover, numerical experiments described in [9] show that the use of a local error control for adaptive time stepsize selection is beneficial for low-dimensional nonlinear Schrödinger equations such as time-dependent Gross–Pitaevskii equations.

Defect-based error estimators. In the present manuscript, our aim is to construct a posteriori local error estimators for higher-order splitting methods applied to linear evolution equations and to analyze them in the context of time-dependent linear Schrödinger equations

$$\begin{cases} \mathrm{i}\,\partial_t \psi(x,t) = -\tfrac{1}{2}\,\Delta\,\psi(x,t) + V(x)\,\psi(x,t)\,, \\ \psi(x,0) = \psi_0(x)\,, \end{cases} \qquad x \in \mathbb{R}^d\,, \quad t \geq 0\,, \tag{1}$$

involving a regular real potential $V : \mathbb{R}^d \to \mathbb{R}$ and a regular initial state $\psi_0 : \mathbb{R}^d \to \mathbb{C}$. Such a local error estimator is a main ingredient in an adaptive time stepsize selection algorithm. In order to expose the technically involved procedure, which extends the construction and error analysis for the particular cases of the first-order Lie–Trotter and the second-order Strang splitting methods, given in our previous work [10], we first focus on a three-stage third-order splitting method and subsequently describe the general approach. We prove asymptotical correctness of the a posteriori local error estimators under natural commutator bounds on the involved operators. Along the way we also recover the known (non)stiff order conditions and a priori convergence bounds, see for example [1, 5, 8]; however, in the present work, we employ an alternative approach based on defects associated with splitting methods, which is also essential for the construction of a posteriori local error estimators. We confirm the theoretical a posteriori local

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error bounds by numerical examples for a test problem of Schrödinger type and also illustrate the error behavior of the a posteriori local error estimators for a test problem of parabolic type.

Extension to nonlinear problems. As in [10], we restrict ourselves to the study of linear evolution equations. The even more technically involved construction and analysis of defect-based local error estimates for nonlinear problems based on the formal calculus of Lie-derivatives will be the subject of future research.

Outline. The structure of the present manuscript is as follows. In Section 2, we state the defect-based local error estimator associated with a higher-order exponential operator splitting method; as our approach is conceptually rather general and not particularly focussed on partial differential equations of Schrödinger type, we employ an abstract framework of evolution equations on Banach spaces. Auxiliary notations and results are collected in Section 3. The construction and analysis of the defect-based local error estimator is carried out in Sections 4 and 5; in order to demonstrate the general procedure with a reasonable amount of involved technicalities, we first focus on a three-stage third-order splitting method and only indicate the extension to higher-order splitting methods. The main tools for a generalization to exponential operator splitting methods of arbitrary order are then explicated in Section 6. In Section 7, in the context of time-dependent linear Schrödinger equations with sufficiently regular problem data we state a result ensuring the asymptotical correctness of the a posteriori local error estimator under natural commutator bounds on the involved operators. Numerical examples for higher-order schemes proposed in the literature [11, 12], given in Section 8, illustrate the error behavior of time-splitting methods for initial-value problems of Schrödinger and parabolic type and in particular confirm the asymptotical correctness of the obtained a posteriori local error estimators.

2. Defect-based error estimators for high-order splitting methods

Linear evolution equation. In the following, we consider the abstract initial value problem

$$\begin{cases} \frac{\mathrm{d}}{\mathrm{d}t}\, u(t) = H\, u(t) = A\, u(t) + B\, u(t)\,, \quad t \geq 0\,, \\ u(0) = u_0\,, \end{cases} \tag{2a}$$

involving the unbounded linear operators $A : D(A) \to \mathcal{B}$, $B : D(B) \to \mathcal{B}$, and $H : D(H) \to \mathcal{B}$, with domains $D(A), D(B), D(H) \subset \mathcal{B}$ such that $\emptyset \neq D(A) \cap D(B) \subset D(H)$ contained in the underlying Banach space $(\mathcal{B}, \|\cdot\|_{\mathcal{B}})$. Due to linearity, it is sufficient to consider the evolution operator associated with (2a)

$$\mathcal{E}(t) = \mathrm{e}^{tH} = \mathrm{e}^{t(A+B)}\,, \quad t \geq 0\,, \tag{2b}$$

which satisfies the initial value problem

$$\begin{cases} \frac{\mathrm{d}}{\mathrm{d}t}\mathcal{E}(t) = H\,\mathcal{E}(t) = A\,\mathcal{E}(t) + B\,\mathcal{E}(t)\,, \quad t \geq 0\,, \\ \mathcal{E}(0) = I\,. \end{cases} \tag{2c}$$

For a particular application the regularity requirements on the initial state are specified later on in Section 5.

High-order splitting methods. For the time integration of (2a) we study an s-stage exponential operator splitting method of (nonstiff) order $p \geq 1$, defined by coefficients $(a_j, b_j)_{j=1}^{s}$. For our purposes, it is useful to consider the numerical evolution operator

$$\begin{gathered} \mathcal{S}(t) = \prod_{j=1}^{s} \mathcal{S}_j(t) = \mathcal{S}_s(t) \cdots \mathcal{S}_1(t) \;\approx\; \mathcal{E}(t)\,, \quad t \geq 0\,, \\ \mathcal{S}_j(t) = \mathrm{e}^{tB_j}\,\mathrm{e}^{tA_j}\,, \quad A_j = a_j\,A\,, \quad B_j = b_j\,B\,, \quad 1 \leq j \leq s\,, \end{gathered} \tag{3a}$$

as time-dependent operator, contrary to practical realisations, where only the evaluation at discrete times is required. Whenever the evolutionary problem (2) with operator A related to the Laplacian originates from a Schrödinger equation, we impose the coefficients to be real, whereas complex coefficients $a_j \in \mathbb{C}$ with $\Re(a_j) > 0$ for $1 \leq j \leq s$ are considered for equations of parabolic type. Henceforth, we assume the basic consistency conditions

$$\text{OC 1:} \qquad \sum_{j=1}^{s} a_j = 1\,, \quad \sum_{j=1}^{s} b_j = 1\,, \tag{3b}$$

to be satisfied.

Local error and defect. As standard, we define the local error as difference between the numerical and exact evolution operators

$$\mathcal{L}(t) = \mathcal{S}(t) - \mathcal{E}(t)\,, \quad t \geq 0\,. \tag{4a}$$

Inserting the splitting operator into equation (2c) for the exact evolution operator further defines the defect

$$\mathcal{D}(t) = \tfrac{\mathrm{d}}{\mathrm{d}t}\mathcal{S}(t) - H\,\mathcal{S}(t)\,, \quad t \geq 0\,. \tag{4b}$$

As a consequence, due to $\frac{\mathrm{d}}{\mathrm{d}t}\mathcal{L} = \frac{\mathrm{d}}{\mathrm{d}t}(\mathcal{S}-\mathcal{E}) = H\,(\mathcal{S}-\mathcal{E}) + \mathcal{D} = H\,\mathcal{L} + \mathcal{D}$ and $\mathcal{S}(0) = I = \mathcal{E}(0)$, the local error satisfies the initial value problem

$$\begin{cases} \tfrac{\mathrm{d}}{\mathrm{d}t}\mathcal{L}(t) = H\,\mathcal{L}(t) + \mathcal{D}(t)\,, \quad t \geq 0\,, \\ \mathcal{L}(0) = 0\,, \end{cases} \tag{4c}$$

and thus by the variation-of-constant formula (Duhamel's principle) the integral representation

$$\mathcal{L}(t) = \int_0^t \mathrm{e}^{(t-\tau)H}\,\mathcal{D}(\tau)\,\mathrm{d}\tau\,, \quad t \geq 0\,, \tag{4d}$$

relating the local error and the defect is obtained.

Local error expansion and order conditions. A standard approach to derive the (nonstiff) order conditions of splitting methods is to require that certain derivatives of the local error vanish at $t = 0$, i.e.,

$$\tfrac{\mathrm{d}}{\mathrm{d}t}\mathcal{L}(0) = \cdots = \tfrac{\mathrm{d}^p}{\mathrm{d}t^p}\mathcal{L}(0) = 0\,. \tag{5a}$$

In regard to the construction and analysis of a posteriori local error estimators, we follow a different approach based on the equivalent conditions

$$\mathcal{D}(0) = \tfrac{\mathrm{d}}{\mathrm{d}t}\mathcal{D}(0) = \cdots = \tfrac{\mathrm{d}^{p-1}}{\mathrm{d}t^{p-1}}\mathcal{D}(0) = 0\,, \tag{5b}$$

rewritten in an appropriate way; for details, see Sections 4 and 6 below.

Defect-based local error estimators. For the purpose of local error estimation the integral in (4d) is approximated by means of an Hermite quadrature formula of order $p+1$

$$\int_0^t f(\tau)\,\mathrm{d}\tau - \mathcal{Q}_f(t) = \mathscr{O}\big(t^{p+2}\big)\,, \quad t \geq 0\,. \tag{6a}$$

More precisely, in order to construct an asymptotically correct a posteriori local error estimator for an s-stage splitting method (3) of order $p \geq 1$, we choose the quadrature approximation

$$\mathcal{Q}_f(t) = \sum_{\ell=0}^{p-1} \omega_\ell\, t^{\ell+1}\,\tfrac{\mathrm{d}^\ell}{\mathrm{d}t^\ell} f(0) + \tfrac{t}{p+1}\,f(t) \tag{6b}$$

such that it relies on the evaluation of the first $p-1$ derivatives of the integrand f at $\tau = 0$ as well as the evaluation of f at $\tau = t$ and further involves certain uniquely defined weights $(\omega_\ell)_{\ell=0}^{p-1}$. Assuming validity of the order conditions (5b) this eventually yields the representation

$$\begin{aligned}\mathcal{P}(t) &= \tfrac{1}{p+1}\, t\, \mathcal{D}(t)\\ &= \tfrac{1}{p+1}\, t \left(\sum_{k=1}^{s} \Big(\prod_{j=k}^{s} \mathcal{S}_j(t) \Big) A_k \Big(\prod_{j=1}^{k-1} \mathcal{S}_j(t) \Big) - A \Big(\prod_{j=1}^{s} \mathcal{S}_j(t) \Big) \right.\\ &\qquad + \sum_{k=1}^{s-1} \Big(\prod_{j=k+1}^{s} \mathcal{S}_j(t) \Big) B_k\, \mathcal{S}_k(t) \Big(\prod_{j=1}^{k-1} \mathcal{S}_j(t) \Big)\\ &\qquad \left. - (1-b_s)\, B\, \mathcal{S}_s(t) \Big(\prod_{j=1}^{s-1} \mathcal{S}_j(t) \Big) \right)\\ &\approx \mathcal{L}(t)\,, \quad t \geq 0\,.\end{aligned} \tag{7}$$

In Section 5 we particularize this construction for a three-stage third-order splitting method and prove the asymptotical correctness

$$\mathcal{P}(t)\, v - \mathcal{L}(t)\, v = \mathscr{O}\big(t^{p+2}\big) \tag{8}$$

of the obtained a posteriori local error estimator under suitable regularity requirements on the argument v. In Section 6 we describe the extension to the general case, and in Section 7 we infer the resulting regularity requirements for linear Schrödinger equations. Furthermore, in Section 8 we illustrate the asymptotical correctness of the local error estimators by numerical examples for higher-order splitting methods applied to linear evolution equations of Schrödinger and parabolic type.

3. Auxiliary notations and results

In this section, we state auxiliary notations and results that are employed throughout.

3.1. Auxiliary notations

Time-independent operators such as A are written in standard font, and time-dependent operators such as $\mathcal{E}$ in calligraphic font.

Definition of related time-independent operators. In the following, we denote

$$H_j = A_j + B_j \,, \tag{9a}$$

$$\overline{H}_j = \sum_{\ell=1}^{j} H_\ell \,, \quad \text{with} \quad \overline{H}_0 = 0 \,, \; \overline{H}_s = H, \tag{9b}$$

$$\widehat{H}_j = \overline{H}_{j-1} + A_j \,, \tag{9c}$$

for $1 \leq j \leq s$; for instance,

$$\begin{aligned} &\widehat{H}_1 = A_1 \,, \\ &\widehat{H}_2 = H_1 + A_2 = A_1 + B_1 + A_2 \,, \\ &\widehat{H}_3 = H_1 + H_2 + A_3 = A_1 + B_1 + A_2 + B_2 + A_3 \,. \end{aligned} \tag{9d}$$

Iterated commutators and related abbreviations. The commutator of two linear operators K, L is given by

$$[K, L] = K\,L - L\,K \,;$$

clearly, the commutator identity

$$[KL, M] = K[L, M] + [K, M]\,L \tag{10}$$

holds. As standard, iterated commutators are defined by

$$\mathrm{ad}_K^0(X) = X \,, \quad \mathrm{ad}_K^n(X) = [K, \mathrm{ad}_K^{n-1}(X)] \,, \quad n \geq 1 \,.$$

In regard to a suitable expansion of the defect, where certain iterated commutators frequently occur, we employ the abbreviations

$$\begin{aligned} A_j^{[0]} &= A_j \,, \quad B_j^{[0]} = B_j \,, \\ A_j^{[\ell]} = [A_j^{[\ell-1]}, \overline{H}_{j-1}] \,, &\quad B_j^{[\ell]} = [B_j^{[\ell-1]}, \overline{H}_j] \,, \qquad \ell \geq 1 \,, \end{aligned} \tag{11}$$

for $1 \leq j \leq s$.

Values of time derivatives. For values of the k-th-order derivative of a time-dependent function we set $\frac{\mathrm{d}^k}{\mathrm{d}t^k} f(0) = \frac{\mathrm{d}^k}{\mathrm{d}t^k} f(t)\big|_{t=0}$.

3.2. Auxiliary results

Sylvester-type equations. A Sylvester equation naturally occurs when determining the first time derivative of the splitting operator associated with the first-order Lie–Trotter splitting method, see also [10]. The following result collects solution representations for Sylvester-type equations needed for a suitable expansion of the splitting operator associated with higher-order splitting methods; later on, it is applied with $A = A_j$ and $B = B_j$.

Lemma 1. *Let A, B, K denote time-independent operators and $\mathcal{G}$ a time-dependent inhomogeneity. Consider the inhomogeneous Sylvester equation*

$$\begin{cases} \frac{\mathrm{d}}{\mathrm{d}t}\mathcal{X} = \mathcal{X}\,A + B\,\mathcal{X} + \mathcal{G}\,, \\ \mathcal{X}(0) \text{ given}\,. \end{cases} \tag{12}$$

(i) The initial value problem (12) *admits the solution representation*

$$\mathcal{X}(t) = \mathrm{e}^{tB}\,\mathcal{X}(0)\,\mathrm{e}^{tA} + \int_0^t \mathrm{e}^{(t-\tau)B}\,\mathcal{G}(\tau)\,\mathrm{e}^{(t-\tau)A}\,\mathrm{d}\tau\,. \tag{13}$$

(ii) Provided that $\mathcal{X}$ satisfies the Sylvester equation (12)*, the time-dependent operators $\mathcal{U}$ and $\mathcal{V}$, defined by*

$$\mathcal{U}(t) = \mathcal{X}(t)\,K\,, \quad \mathcal{V}(t) = K\,\mathcal{X}(t)\,, \tag{14a}$$

are solutions of the Sylvester equations

$$\begin{cases} \frac{\mathrm{d}}{\mathrm{d}t}\mathcal{U} = \mathcal{U}\,A + B\,\mathcal{U} + \mathcal{X}\,[A,K] + \mathcal{G}\,K\,, \\ \mathcal{U}(0) = \mathcal{X}(0)\,K\,, \end{cases} \tag{14b}$$

and

$$\begin{cases} \frac{\mathrm{d}}{\mathrm{d}t}\mathcal{V} = \mathcal{V}\,A + B\,\mathcal{V} + [K,B]\,\mathcal{X} + K\,\mathcal{G}\,, \\ \mathcal{V}(0) = K\,\mathcal{X}(0)\,, \end{cases} \tag{14c}$$

respectively.

(iii) Provided that $\mathcal{X}$ satisfies the Sylvester equation (12)*, the first commutator $\mathcal{W}(t) = [\mathcal{X}, K](t) = [\mathcal{X}(t), K]$ is the solution of the Sylvester equation*

$$\begin{cases} \frac{\mathrm{d}}{\mathrm{d}t}\mathcal{W} = \mathcal{W}\,A + B\,\mathcal{W} + \mathcal{X}\,[A,K] + [B,K]\,\mathcal{X} + [\,\mathcal{G}, K]\,, \\ \mathcal{W}(0) = [\mathcal{X}(0), K]\,. \end{cases} \tag{15}$$

PROOF. (i) Straightforward verification, see also [10].

(ii) Differentiating $\mathcal{U}$ we obtain from (12)

$$\begin{aligned}\tfrac{\mathrm{d}}{\mathrm{d}t}\mathcal{U} = (\tfrac{\mathrm{d}}{\mathrm{d}t}\mathcal{X})\,K &= \mathcal{X}\,A\,K + B\,\mathcal{X}\,K + \mathcal{G}\,K \\ &= \mathcal{X}\,K\,A + B\,\mathcal{X}\,K + \mathcal{X}\,(A\,K - K\,A) + \mathcal{G}\,K \\ &= \mathcal{U}\,A + B\,\mathcal{U} + \mathcal{X}\,[A,K] + \mathcal{G}\,K\,,\end{aligned}$$

and analogously for $\mathcal{V}$.

(iii) This follows directly from (ii) by combining (14b) and (14c). □

Evolution equation for a triple operator product. In the following, we make use of the fact that the product of time-dependent operators which are solutions to Sylvester equations of the form

$$\begin{cases} \tfrac{\mathrm{d}}{\mathrm{d}t}\mathcal{X}_j = \mathcal{X}_j\,A_j + B_j\,\mathcal{X}_j + \mathcal{G}_j\,, \\ \mathcal{X}_j(0)\ \text{given}\,, \end{cases} \tag{16a}$$

satisfies an evolution equation with dominant part involving H and a certain inhomogeneity. In particular, in Section 4 we employ the relation

$$\begin{aligned}\tfrac{\mathrm{d}}{\mathrm{d}t}\big(\mathcal{X}_3\,\mathcal{X}_2\,\mathcal{X}_1\big) = H\,\mathcal{X}_3\,\mathcal{X}_2\,\mathcal{X}_1 & \\ + \big([\mathcal{X}_3,\widehat{H}_3] + \mathcal{G}_3\big)\,\mathcal{X}_2\,\mathcal{X}_1 & \\ + \mathcal{X}_3\,\big([\mathcal{X}_2,\widehat{H}_2] + \mathcal{G}_2\big)\,\mathcal{X}_1 & \\ + \mathcal{X}_3\,\mathcal{X}_2\,\big([\mathcal{X}_1,\widehat{H}_1] + \mathcal{G}_1\big)\,, &\end{aligned} \tag{16b}$$

obtained for a triple product, provided that (3b) holds with $s = 3$; this is verified by a straightforward calculation or follows from Lemma 2 deduced in Section 6.

Further notation and preliminary remarks. For the sake of compact and consistent notations, we introduce

$$\delta\mathcal{X} = \tfrac{\mathrm{d}}{\mathrm{d}t}\mathcal{X} - H\mathcal{X}\,, \tag{17}$$

and define by recurrence

$$\begin{gathered}\mathcal{S}_j^{(0)}(t) = e^{tB_j}\,e^{tA_j}\,,\quad 1 \le j \le s\,,\qquad \mathcal{S}^{(0)} = \mathcal{S}_s^{(0)}\,\cdots\,\mathcal{S}_1^{(0)}\,,\\ \mathcal{S}^{(n)} = \delta\mathcal{S}^{(n-1)}\,,\quad n \ge 1\,.\end{gathered} \tag{18}$$

Clearly, it holds $\mathcal{S}_j^{(0)} = \mathcal{S}_j$ and $\mathcal{S}^{(0)} = \mathcal{S}$. We note that the defect (4b) equals

$$\mathcal{D} = \tfrac{\mathrm{d}}{\mathrm{d}t}\mathcal{S} - H\mathcal{S} = \delta\mathcal{S}^{(0)} = \mathcal{S}^{(1)} \,. \tag{19}$$

In order to capture the residual of a time-dependent operator $\mathcal{X}$ with respect to the j-th homogeneous Sylvester equation, we set

$$\sigma_j(\mathcal{X}) = \tfrac{\mathrm{d}}{\mathrm{d}t}\mathcal{X} - \mathcal{X} A_j - B_j\,\mathcal{X}\,, \quad 1 \leq j \leq s\,; \tag{20}$$

thus, identity (16b) can be reformulated as

$$\begin{aligned}\delta\left(\mathcal{X}_3\,\mathcal{X}_2\,\mathcal{X}_1\right) = \big([\mathcal{X}_3, \widehat{H}_3] + \sigma_3(\mathcal{X}_3)\big)\,\mathcal{X}_2\,\mathcal{X}_1 \\ + \mathcal{X}_3\left([\mathcal{X}_2, \widehat{H}_2] + \sigma_2(\mathcal{X}_2)\right)\mathcal{X}_1 \\ + \mathcal{X}_3\,\mathcal{X}_2\left([\mathcal{X}_1, \widehat{H}_1] + \sigma_1(\mathcal{X}_1)\right).\end{aligned} \tag{21}$$

Inserting $\mathcal{X}_j = \mathcal{S}_j^{(0)}$ into (21) and noting that $\sigma_j\big(\mathcal{S}_j^{(0)}\big) = 0$ motivates the abbreviation $\mathcal{S}_j^{(1)} = [\mathcal{S}_j^{(0)}, \widehat{H}_j] + \sigma_j\big(\mathcal{S}_j^{(0)}\big) = [\mathcal{S}_j^{(0)}, \widehat{H}_j]$; more generally, we define

$$\mathcal{S}_j^{(k)} = [\mathcal{S}_j^{(k-1)}, \widehat{H}_j] + \sigma_j\big(\mathcal{S}_j^{(k-1)}\big)\,, \quad k \geq 1\,. \tag{22}$$

4. A priori local error analysis

Objective. In this section, our objective is to provide a suitable expansion of the local error (4a) ensuring

$$\mathcal{L}(t)\,v = \mathscr{O}\big(t^{p+1}\big)$$

under certain regularity requirements on the argument v. By the integral relation (4d) and due to (19) this is equivalent to

$$\mathcal{D}(t)\,v = \mathcal{S}^{(1)}(t)\,v = \mathscr{O}\big(t^{p}\big)\,,$$

provided that the exact evolution operator remains bounded on the underlying function space.

Approach. In principle, an appropriate expansion of the defect could be obtained by a standard Taylor series expansion

$$\mathcal{D}(t) = \mathcal{S}^{(1)}(t) = \sum_{\ell=0}^{p-1} \tfrac{1}{\ell!}\, t^{\ell}\, \tfrac{\mathrm{d}^{\ell}}{\mathrm{d}t^{\ell}}\, \mathcal{S}^{(1)}(0) + \int_{T_p} \tfrac{\mathrm{d}^{p}}{\mathrm{d}t^{p}}\, \mathcal{S}^{(1)}(\tau_p)\, \mathrm{d}\tau\,,$$
$$T_p = \{\tau = (\tau_1, \dots, \tau_p) \in \mathbb{R}^p : 0 \leq \tau_p \leq \cdots \leq \tau_1 \leq t\}\,,$$

where the p-th-order conditions on the coefficients of the splitting method correspond to the conditions

$$\tfrac{\mathrm{d}^{\ell}}{\mathrm{d}t^{\ell}}\, \mathcal{S}^{(1)}(0) = 0\,, \quad 0 \leq \ell \leq p-1\,, \tag{23}$$

which are also utilized in the construction of the a posteriori local error estimator. However, for a method of higher order involving a higher number of stages this approach becomes unfeasible, due to the rapidly increasing number of terms involved in $\tfrac{\mathrm{d}^{p}}{\mathrm{d}t^{p}}\, \mathcal{S}^{(1)}$. We note that in addition a careful inspection of $\tfrac{\mathrm{d}^{p}}{\mathrm{d}t^{p}}\, \mathcal{S}^{(1)}$ and a suitable reformulation of the involved operators as iterated commutators is required in order to retain the optimal regularity requirements on the argument v.

Alternative approach. In this work, we follow a different approach based on the derivation of suitable differential equations for the splitting operator, the defect and its higher derivatives. In this section, we expound our approach for a three-stage third-order splitting method, where a stepwise expansion of part of the integrand in (4d) yields the multiple integral representation

$$\begin{aligned} p = 3: \qquad \mathcal{L}(t) &= \int_0^t \mathrm{e}^{(t-\tau_1)H}\, \mathcal{S}^{(1)}(\tau_1)\, \mathrm{d}\tau_1 \\ &= \int_0^t \int_0^{\tau_1} \mathrm{e}^{(t-\tau_2)H}\, \mathcal{S}^{(2)}(\tau_2)\, \mathrm{d}\tau_2\, \mathrm{d}\tau_1 \\ &= \int_0^t \int_0^{\tau_1} \int_0^{\tau_2} \mathrm{e}^{(t-\tau_3)H}\, \mathcal{S}^{(3)}(\tau_3)\, \mathrm{d}\tau_3\, \mathrm{d}\tau_2\, \mathrm{d}\tau_1\,, \end{aligned} \tag{24a}$$

provided that the imposed order conditions (23)

$$p = 3: \qquad \mathcal{S}^{(1)}(0) = 0\,, \quad \tfrac{\mathrm{d}}{\mathrm{d}t}\mathcal{S}^{(1)}(0) = 0\,, \quad \tfrac{\mathrm{d}^2}{\mathrm{d}t^2}\, \mathcal{S}^{(1)}(0) = 0\,, \tag{24b}$$

hold. In the following subsections, the derivation of the expansion (24a) is explicated in detail, and, as a consequence, the desired a priori local error expansion $\mathcal{L}(t)\, v = \mathscr{O}(t^4)$ is retained under certain regularity requirements on the argument v, see Proposition 1 below. For a p-th-order splitting method the same procedure leads to the following local error expansion

$$\begin{aligned} \mathcal{L}(t) &= \int_{T_p} \mathrm{e}^{(t-\tau_p)H}\, \mathcal{S}^{(p)}(\tau_p)\, \mathrm{d}\tau\,, \\ T_p &= \{\tau = (\tau_1, \dots, \tau_p) \in \mathbb{R}^p : 0 \le \tau_p \le \dots \le \tau_1 \le t\}\,, \end{aligned} \tag{25}$$

provided that the order conditions (23) are satisfied. Our approach establishes an explicit representation for the local error, which, however, is of high complexity for higher-order splitting methods. As it suffices to investigate the structure of the terms involved, we refrain from a specification of the resulting local error representation.

4.1. A first representation for the defect

For a splitting method involving three stages, i.e. $s = 3$, the defining relation (4b) for the defect reduces to

$$\mathcal{S}^{(1)} = \delta \mathcal{S}^{(0)}\,, \quad \mathcal{S}^{(0)} = \mathcal{S}_3^{(0)}\, \mathcal{S}_2^{(0)}\, \mathcal{S}_1^{(0)}\,, \tag{26a}$$

see also (18) and (19). Observing that $\mathcal{S}_j^{(0)}$ satisfies the initial value problem

$$\begin{cases} \frac{\mathrm{d}}{\mathrm{d}t} \mathcal{S}_j^{(0)} = \mathcal{S}_j^{(0)} A_j + B_j\, \mathcal{S}_j^{(0)}\,, \\ \mathcal{S}_j^{(0)}(0) = I\,, \end{cases} \tag{26b}$$

relation (21) (with $\mathcal{X}_j = \mathcal{S}_j^{(0)}$ and $\sigma_j\big(\mathcal{S}_j^{(0)}\big) = 0$) together with the notation $\mathcal{S}_j^{(1)} = [\mathcal{S}_j^{(0)}, \widehat{H}_j]$, see (22), yields the following initial value problem for the splitting operator,

$$\begin{cases} \delta \mathcal{S}^{(0)} = \mathcal{S}_3^{(1)}\, \mathcal{S}_2^{(0)}\, \mathcal{S}_1^{(0)} + \mathcal{S}_3^{(0)}\, \mathcal{S}_2^{(1)}\, \mathcal{S}_1^{(0)} + \mathcal{S}_3^{(0)}\, \mathcal{S}_2^{(0)}\, \mathcal{S}_1^{(1)}\,, \\ \mathcal{S}^{(0)}(0) = I\,. \end{cases} \tag{26c}$$

We note that this also provides a representation for the defect, namely,

$$\mathcal{S}^{(1)} = \mathcal{S}_3^{(1)}\, \mathcal{S}_2^{(0)}\, \mathcal{S}_1^{(0)} + \mathcal{S}_3^{(0)}\, \mathcal{S}_2^{(1)}\, \mathcal{S}_1^{(0)} + \mathcal{S}_3^{(0)}\, \mathcal{S}_2^{(0)}\, \mathcal{S}_1^{(1)}\,, \tag{27}$$

see (26a). The obvious generalization to an s-stage splitting method is

$$\mathcal{S}^{(1)} = \sum_{k_1 + \dots + k_s = 1} \mathcal{S}_s^{(k_s)} \cdots \mathcal{S}_1^{(k_1)}\,.$$

4.2. A first expansion step ensuring $\mathcal{L}(t)\,v = \mathscr{O}(t^2)$

Aim. Our starting point is the local error representation given above,

$$\begin{aligned}\mathcal{L}(t) &= \int_0^t \mathrm{e}^{(t-\tau_1)H}\,\mathcal{S}^{(1)}(\tau_1)\,\mathrm{d}\tau_1\,,\\ \mathcal{S}^{(1)} &= \mathcal{S}_3^{(1)}\,\mathcal{S}_2^{(0)}\,\mathcal{S}_1^{(0)} + \mathcal{S}_3^{(0)}\,\mathcal{S}_2^{(1)}\,\mathcal{S}_1^{(0)} + \mathcal{S}_3^{(0)}\,\mathcal{S}_2^{(0)}\,\mathcal{S}_1^{(1)}\,,\end{aligned} \tag{28}$$

ensuring $\mathcal{L}(t)\,v = \mathscr{O}(t)$ under certain regularity assumptions on v, see also (4d), (22), and (27). In a first step, we aim for a suitable integral representation for the terms $\mathcal{S}_j^{(1)}$ such that even $\mathcal{L}(t)\,v = \mathscr{O}(t^2)$.

Integral representations for $\mathcal{S}_j^{(1)}$. For convenience, we recall the abbreviations $\mathcal{S}_j^{(0)}(t) = e^{tB_j}\,e^{tA_j}$, $\mathcal{S}_j^{(1)} = [\mathcal{S}_j^{(0)}, \widehat{H}_j]$, and $A_j^{[1]} = [A_j, \overline{H}_{j-1}] = [A_j, \widehat{H}_j]$ as well as $B_j^{[1]} = [B_j, \overline{H}_j] = [B_j, \widehat{H}_j]$. Relation (15) implies

$$\begin{cases} \sigma\big(\mathcal{S}_j^{(1)}\big) = \frac{\mathrm{d}}{\mathrm{d}t}\mathcal{S}_j^{(1)} - \mathcal{S}_j^{(1)} A_j - B_j\,\mathcal{S}_j^{(1)} = \mathcal{S}_j^{(0)}\,A_j^{[1]} + B_j^{[1]}\,\mathcal{S}_j^{(0)}\,,\\ \mathcal{S}_j^{(1)}(0) = 0\,, \end{cases} \tag{29}$$

see also (20), and thus an application of the variation-of-constants formula (13)

$$\mathcal{S}_j^{(1)}(t) = \int_0^t \mathrm{e}^{(t-\tau)B_j}\,\big(\mathcal{S}_j^{(0)}(\tau)\,A_j^{[1]} + B_j^{[1]}\,\mathcal{S}_j^{(0)}(\tau)\big)\,\mathrm{e}^{(t-\tau)A_j}\,\mathrm{d}\tau \tag{30}$$

ensures $\mathcal{S}_j^{(1)}(t)\,v = \mathscr{O}(t)$.

Local error expansion. Inserting the integral representation (30) into (28) shows $\mathcal{L}(t)\,v = \mathscr{O}(t^2)$. In the context of linear Schrödinger equations, where the operator A is related to the Laplacian and B to a smooth potential, it is seen that $\mathcal{S}_j^{(1)}(t)\,v$ and as a consequence $\mathcal{L}(t)\,v$ are well-defined in the Lebesgue space L^2 for arguments v in the Sobolev space H^1, see Section 7 for further details. Generally, the necessary assumption will reduce to a regularity requirement on the exact solution of the underlying differential equation.

4.3. A further expansion step ensuring $\mathcal{L}(t)\,v = \mathscr{O}(t^3)$

Aim. In order to expand the local error further, we revisit (28) and deduce an initial value problem for the defect $\mathcal{S}^{(1)}$, aiming for an integral representation of the form

$$\mathcal{S}^{(1)}(\tau_1) = \mathrm{e}^{\tau_1 H} \mathcal{S}^{(1)}(0) + \int_0^{\tau_1} \mathrm{e}^{(\tau_1 - \tau_2) H}\, \mathcal{S}^{(2)}(\tau_2)\, \mathrm{d}\tau_2\,. \tag{31}$$

Due to the validity of the first-order conditions (3b), which correspond to

$$\text{OC 1:} \qquad \mathcal{S}^{(1)}(0) = 0 \quad \Longleftrightarrow \quad \sum_{j=1}^{s} a_j = 1, \quad \sum_{j=1}^{s} b_j = 1\,, \tag{32}$$

this further leads to

$$\mathcal{L}(t) = \int_0^t \int_0^{\tau_1} \mathrm{e}^{(t - \tau_2) H}\, \mathcal{S}^{(2)}(\tau_2)\, \mathrm{d}\tau_2\, \mathrm{d}\tau_1\,. \tag{33}$$

Utilizing an integral representation for building blocks constituting $\mathcal{S}^{(2)}$ it turns out that even $\mathcal{L}(t)\,v = \mathscr{O}(t^3)$, provided that an additional order condition is satisfied.

Initial value problem for $\mathcal{S}^{(1)}$. We recall (29)

$$\begin{cases} \sigma_j\big(\mathcal{S}_j^{(1)}\big) = \mathcal{S}_j^{(0)} A_j^{[1]} + B_j^{[1]}\, \mathcal{S}_j^{(0)}\,, \\ \mathcal{S}_j^{(1)}(0) = 0\,, \end{cases}$$

and that the defect is given by

$$\mathcal{S}^{(1)} = \mathcal{S}_3^{(1)}\, \mathcal{S}_2^{(0)}\, \mathcal{S}_1^{(0)} + \mathcal{S}_3^{(0)}\, \mathcal{S}_2^{(1)}\, \mathcal{S}_1^{(0)} + \mathcal{S}_3^{(0)}\, \mathcal{S}_2^{(0)}\, \mathcal{S}_1^{(1)}\,,$$

see (27). Relation (21) applied for instance to the triple product $\mathcal{S}_3^{(1)}\, \mathcal{S}_2^{(0)}\, \mathcal{S}_1^{(0)}$ (setting $\mathcal{X}_3 = \mathcal{S}_3^{(1)}$ as well as $\mathcal{X}_j = \mathcal{S}_j^{(0)}$ for $j = 1, 2$ and utilizing $\sigma_j(\mathcal{S}_j^{(0)}) = 0$ for $j = 1, 2$) implies

$$\begin{aligned} \delta\big(\mathcal{S}_3^{(1)}\, \mathcal{S}_2^{(0)}\, \mathcal{S}_1^{(0)}\big) &= \big([\mathcal{S}_3^{(1)}, \widehat{H}_3] + \sigma_3(\mathcal{S}_3^{(1)})\big)\, \mathcal{S}_2^{(0)}\, \mathcal{S}_1^{(0)} \\ &\qquad + \mathcal{S}_3^{(1)}\, [\mathcal{S}_2^{(0)}, \widehat{H}_2]\, \mathcal{S}_1^{(0)} \\ &\qquad\quad + \mathcal{S}_3^{(1)}\, \mathcal{S}_2^{(0)}\, [\mathcal{S}_1^{(0)}, \widehat{H}_1] \\ &= \mathcal{S}_3^{(2)}\, \mathcal{S}_2^{(0)}\, \mathcal{S}_1^{(0)} + \mathcal{S}_3^{(1)}\, \mathcal{S}_2^{(1)}\, \mathcal{S}_1^{(0)} + \mathcal{S}_3^{(1)}\, \mathcal{S}_2^{(0)}\, \mathcal{S}_1^{(1)}\,, \end{aligned}$$

where $\mathcal{S}_j^{(2)} = [\mathcal{S}_j^{(1)}, \widehat{H}_j] + \sigma_j(\mathcal{S}_j^{(1)})$, see (22); clearly, $\delta\big(\mathcal{S}_3^{(0)}\,\mathcal{S}_2^{(1)}\,\mathcal{S}_1^{(0)}\big)$ and $\delta\big(\mathcal{S}_3^{(0)}\,\mathcal{S}_2^{(0)}\,\mathcal{S}_1^{(1)}\big)$ can be represented in an analogous way. By summation we thus obtain an initial value problem for the defect, rewritten as

$$\begin{cases} \delta\mathcal{S}^{(1)} = \mathcal{S}^{(2)}\,, \\ \mathcal{S}^{(1)}(0) = 0\,, \end{cases} \tag{34}$$

involving the inhomogeneity

$$\begin{aligned} \mathcal{S}^{(2)} = \quad & \mathcal{S}_3^{(2)}\,\mathcal{S}_2^{(0)}\,\mathcal{S}_1^{(0)} + \mathcal{S}_3^{(1)}\,\mathcal{S}_2^{(1)}\,\mathcal{S}_1^{(0)} + \mathcal{S}_3^{(1)}\,\mathcal{S}_2^{(0)}\,\mathcal{S}_1^{(1)} \\ & + \mathcal{S}_3^{(1)}\,\mathcal{S}_2^{(1)}\,\mathcal{S}_1^{(0)} + \mathcal{S}_3^{(0)}\,\mathcal{S}_2^{(2)}\,\mathcal{S}_1^{(0)} + \mathcal{S}_3^{(0)}\,\mathcal{S}_2^{(1)}\,\mathcal{S}_1^{(1)} \\ & + \mathcal{S}_3^{(1)}\,\mathcal{S}_2^{(0)}\,\mathcal{S}_1^{(1)} + \mathcal{S}_3^{(0)}\,\mathcal{S}_2^{(1)}\,\mathcal{S}_1^{(1)} + \mathcal{S}_3^{(0)}\,\mathcal{S}_2^{(0)}\,\mathcal{S}_1^{(2)}\,, \end{aligned} \tag{35}$$

where $\mathcal{S}_j^{(0)}(t)\,v = \mathscr{O}(1)$ and $\mathcal{S}_j^{(1)}(t)\,v = \mathscr{O}(t)$. It remains to investigate the leading contributions involving $S_j^{(2)}$.

Generalization. Again, it is straightforward to extend the above considerations to an s-stage splitting method, which leads to the representation

$$\mathcal{S}^{(2)} = \sum_{k_1+\cdots+k_s=2} \tfrac{2!}{k_1!\cdots k_s!}\,\mathcal{S}_s^{(k_s)}\cdots\mathcal{S}_1^{(k_1)}\,,$$

see Section 6.

Integral representations for $\mathcal{S}_j^{(2)}$ and structure of the term $\mathcal{S}^{(2)}$. In order to ensure $\mathcal{L}(t)\,v = \mathscr{O}(t^3)$, we next deduce an integral representation for the quantities $\mathcal{S}_j^{(2)} = [\mathcal{S}_j^{(1)}, \widehat{H}_j] + \sigma_j\big(\mathcal{S}_j^{(1)}\big)$, see also (22); this step is accomplished by invoking Lemma 4 with $k = 2$, see Section 6. An application of relation (59b) shows that $\mathcal{S}_j^{(2)}$ satisfies an initial value problem, rewritten as

$$\begin{cases} \sigma_j\big(\mathcal{S}_j^{(2)}\big) = 2\left(\mathcal{S}_j^{(1)} A_j^{[1]} + B_j^{[1]}\,\mathcal{S}_j^{(1)}\right) + \mathcal{S}_j^{(0)} A_j^{[2]} + B_j^{[2]}\,\mathcal{S}_j^{(0)}\,, \\ \mathcal{S}_j^{(2)}(0) = A_j^{[1]} + B_j^{[1]}\,; \end{cases} \tag{36}$$

involving the second iterated commutators

$$A_j^{[2]} = [[A_j, \overline{H}_{j-1}], \overline{H}_{j-1}], \quad B_j^{[2]} = [[B_j, \overline{H}_j], \overline{H}_j]\,;$$

recalling that $\mathcal{S}_j^{(0)}(0) = I$ and $\mathcal{S}_j^{(1)}(0) = 0$, the relation for the initial value is obtained from the identity $\mathcal{S}_j^{(2)}(0) = \sigma_j(\mathcal{S}_j^{(1)})(0) = A_j^{[1]} + B_j^{[1]}$, see (29). As a consequence, the integral representation

$$\begin{aligned}\mathcal{S}_j^{(2)}(t) &= \mathrm{e}^{tB_j}\,\big(A_j^{[1]} + B_j^{[1]}\big)\,\mathrm{e}^{tA_j} \\ &\qquad + 2\int_0^t \mathrm{e}^{(t-\tau)B_j}\,\big(\mathcal{S}_j^{(1)}(\tau)\,A_j^{[1]} + B_j^{[1]}\,\mathcal{S}_j^{(1)}(\tau)\big)\,\mathrm{e}^{(t-\tau)A_j}\,\mathrm{d}\tau \\ &\qquad\quad + \int_0^t \mathrm{e}^{(t-\tau)B_j}\,\big(\mathcal{S}_j^{(0)}(\tau)\,A_j^{[2]} + B_j^{[2]}\,\mathcal{S}_j^{(0)}(\tau)\big)\,\mathrm{e}^{(t-\tau)A_j}\,\mathrm{d}\tau\,,\end{aligned} \tag{37}$$

is obtained, where $\mathcal{S}_j^{(1)}$ is expressed by (30). We point out that the first term in (37) satisfies $\mathrm{e}^{tB_j}\,\big(A_j^{[1]} + B_j^{[1]}\big)\,\mathrm{e}^{tA_j}\,v = \mathscr{O}(1)$ only. In order to ensure that the leading term in $\mathcal{S}^{(2)}$, given by

$$\begin{aligned}\mathcal{S}^{(2;0)}(t) &= \mathrm{e}^{tB_3}\,\big(A_3^{[1]} + B_3^{[1]}\big)\,\mathrm{e}^{tA_3}\,\mathcal{S}_2^{(0)}(t)\,\mathcal{S}_1^{(0)}(t) \\ &\qquad + \mathcal{S}_3^{(0)}(t)\,\mathrm{e}^{tB_2}\,\big(A_2^{[1]} + B_2^{[1]}\big)\,\mathrm{e}^{tA_2}\,\mathcal{S}_1^{(0)}(t) \\ &\qquad\quad + \mathcal{S}_3^{(0)}(t)\,\mathcal{S}_2^{(0)}(t)\,\mathrm{e}^{tB_1}\,\big(A_1^{[1]} + B_1^{[1]}\big)\,\mathrm{e}^{tA_1}\,,\end{aligned}$$

satisfies $\mathcal{S}^{(2;0)}(t) = \mathscr{O}(t)$, we employ the second-order condition

$$\begin{aligned}\text{OC 2:}\qquad & \mathcal{S}^{(2)}(0) = \sum_{j=1}^{s}\big(A_j^{[1]} + B_j^{[1]}\big) = 0 \\ & \iff\quad \sum_{j=1}^{s}\sum_{k=1}^{j-1} a_j b_k - \sum_{j=1}^{s}\sum_{k=1}^{j} b_j a_k = 0\,,\end{aligned} \tag{38}$$

which in particular implies $\mathcal{S}^{(2;0)}(0) = 0$; recall that $\mathcal{S}^{(0)}(0) = I$. By means of the variation-of-constants formula

$$\mathcal{S}^{(2;0)}(t) = \int_0^t \mathrm{e}^{(t-\tau)H}\,\delta\mathcal{S}^{(2;0)}(\tau)\,\mathrm{d}\tau\,,$$

this further yields $\mathcal{S}^{(2;0)}(t)\,v = \mathscr{O}(t)$, as desired. Thus, together with the integral representation (30) the relation $\mathcal{S}^{(2)}(t) = \mathscr{O}(t)$ readily follows. It remains to specify the structure of the obtained representation for $\mathcal{S}^{(2)}$ and in particular of $\delta\mathcal{S}^{(2;0)}$. Due to $\sigma_j\big(\mathcal{S}_j^{(0)}\big) = 0$, and with

$$\mathcal{X}_j(t) = \mathrm{e}^{tB_j}\big(A_j^{[1]} + B_j^{[1]}\big)\,\mathrm{e}^{tA_j}\,,\quad \mathcal{X}_j(0) = A_j^{[1]} + B_j^{[1]}\,,\quad \sigma_j\big(\mathcal{X}_j\big) = 0\,,$$

relation (21) yields

$$
\begin{aligned}
\delta\mathcal{S}^{(2;0)} = [\mathcal{X}_3, \widehat{H}_3]\, \mathcal{S}_2^{(0)}\, \mathcal{S}_1^{(0)} + \mathcal{X}_3\, \mathcal{S}_2^{(1)}\, \mathcal{S}_1^{(0)} + \mathcal{X}_3\, \mathcal{S}_2^{(0)}\, \mathcal{S}_1^{(1)} \\
+ \mathcal{S}_3^{(1)}\, \mathcal{X}_2\, \mathcal{S}_1^{(0)} + \mathcal{S}_3^{(0)}\, [\mathcal{X}_2, \widehat{H}_2]\, \mathcal{S}_1^{(0)} + \mathcal{S}_3^{(0)}\, \mathcal{X}_2\, \mathcal{S}_1^{(1)} \\
+ \mathcal{S}_3^{(1)}\, \mathcal{S}_2^{(0)}\, \mathcal{X}_1 + \mathcal{S}_3^{(0)}\, \mathcal{S}_2^{(1)}\, \mathcal{X}_1 + \mathcal{S}_3^{(0)}\, \mathcal{S}_2^{(0)}\, [\mathcal{X}_1, \widehat{H}_1]\,.
\end{aligned}
$$

An application of Lemma 1 (iii) and (i) further implies

$$
\begin{aligned}
\sigma_j([\mathcal{X}_j, \widehat{H}_j]) &= \mathcal{X}_j\, A_j^{[1]} + B_j^{[1]}\, \mathcal{X}_j\,, \\
[\mathcal{X}_j, \widehat{H}_j](t) &= \mathrm{e}^{tB_j}\, \big(A_j^{[1]} + B_j^{[1]}\big)\, \mathrm{e}^{tA_j} \\
&\quad + \int_0^t \mathrm{e}^{(t-\tau)B_j}\, \big(\mathcal{X}_j(\tau)\, A_j^{[1]} + B_j^{[1]}\, \mathcal{X}_j(\tau)\big)\, \mathrm{e}^{(t-\tau)A_j}\; \mathrm{d}\tau\,.
\end{aligned}
$$

This shows that $\delta\mathcal{S}^{(2;0)}$ can be expressed via compositions of at most two commutators and evolution operators associated with the subproblems. Similarly, the remaining contributions to $\mathcal{S}^{(2)}$ involving $\mathcal{S}_j^{(1)}$ are of this structure, see (35) and (30). Altogether, this implies that the quantity $\mathcal{S}^{(2)}$ comprises compositions of at most two commutators and evolution operators.

Local error expansion. Subsuming the above considerations concerning $\mathcal{S}^{(2)}$ and inserting the obtained integral representations for $\mathcal{S}_j^{(1)}$ and $\mathcal{S}_j^{(2)}$ into (33) leads to a local error expansion implying $\mathcal{L}(t)\, v = \mathscr{O}(t^3)$. In the context of linear Schrödinger equations the term $\mathcal{S}^{(2)}\, v$ and thus $\mathcal{L}(t)\, v$ is well-defined in L^2 for $v \in H^2$, see also Section 7.

4.4. A final expansion step ensuring $\mathcal{L}(t)\, v = \mathscr{O}(t^4)$

Aim. We employ the same procedure as before, with some additional technicalities. Revisiting formula (33) and applying the second-order condition $\mathcal{S}^{(2)}(0) = 0$, see (38), our aim is to deduce an initial value problem for $\mathcal{S}^{(2)}$ in order to obtain an integral representation of the form

$$
\mathcal{S}^{(2)}(\tau_2) = \int_0^{\tau_2} \mathrm{e}^{(\tau_2-\tau_3)H}\, \mathcal{S}^{(3)}(\tau_3)\; \mathrm{d}\tau_3\,, \tag{39}
$$

which leads to

$$
\mathcal{L}(t) = \int_0^t \int_0^{\tau_1} \int_0^{\tau_2} \mathrm{e}^{(t-\tau_3)H}\, \mathcal{S}^{(3)}(\tau_3)\; \mathrm{d}\tau_3\, \mathrm{d}\tau_2\, \mathrm{d}\tau_1\,. \tag{40}
$$

Similarly as before, with the help of suitable integral representations for building blocks constituting $\mathcal{S}^{(3)}$ it turns out that even $\mathcal{L}(t)\, v = \mathscr{O}(t^4)$, provided that the third-order conditions are satisfied.

Initial value problem for $\mathcal{S}^{(2)}$. In order to obtain a further expansion of the term $\mathcal{S}^{(2)}$, we invoke Lemma 3 given in Section 6 with $s = 3$ and $k = 3$

$$\mathcal{S}^{(3)} = \delta \mathcal{S}^{(2)} = \sum_{k_1+k_2+k_3=3} \tfrac{3!}{k_1!\,k_2!\,k_3!}\, \mathcal{S}_3^{(k_3)}\, \mathcal{S}_2^{(k_2)}\, \mathcal{S}_1^{(k_1)}\,, \tag{41}$$

with $\mathcal{S}_j^{(k)}$ defined by the recurrence in (22). That is, the operator $\mathcal{S}^{(2)}$ satisfies the initial value problem

$$\begin{cases} \delta \mathcal{S}^{(2)} = \mathcal{S}^{(3)}\,, \\ \mathcal{S}^{(2)}(0) = 0\,, \end{cases} \tag{42}$$

involving the inhomogeneity $\mathcal{S}^{(3)}$, see also (41).

Generalization. It is straightforward to extend the above considerations to a splitting method involving s stages, yielding

$$\mathcal{S}^{(3)} = \sum_{k_1+\cdots+k_s=3} \tfrac{3!}{k_1!\cdots k_s!}\, \mathcal{S}_s^{(k_s)} \cdots \mathcal{S}_1^{(k_1)}\,,$$

see Section 6.

Integral representations for $\mathcal{S}_j^{(3)}$ and structure of the term $\mathcal{S}^{(3)}$. In order to ensure $\mathcal{L}(t)\,v = \mathscr{O}(t^4)$, we deduce an integral representation for the quantities $\mathcal{S}_j^{(3)}$. Invoking again Lemma 4 with $k = 3$ implies

$$\begin{aligned} \sigma\big(\mathcal{S}_j^{(3)}\big) &= \tfrac{\mathrm{d}}{\mathrm{d}t} \mathcal{S}_j^{(3)} - \mathcal{S}_j^{(3)} A_j - B_j\, \mathcal{S}_j^{(3)} \\ &= 3\,\big(\mathcal{S}_j^{(2)} A_j^{[1]} + B_j^{[1]}\, \mathcal{S}_j^{(2)}\big) \\ &\quad + 3\,\big(\mathcal{S}_j^{(1)} A_j^{[2]} + B_j^{[2]}\, \mathcal{S}_j^{(1)}\big) \\ &\quad + \mathcal{S}_j^{(0)} A_j^{[3]} + B_j^{[3]}\, \mathcal{S}_j^{(0)}\,, \end{aligned}$$

involving the third iterated commutators

$$A_j^{[3]} = [[[A_j, \overline{H}_{j-1}], \overline{H}_{j-1}], \overline{H}_{j-1}]\,, \quad B_j^{[3]} = [[[B_j, \overline{H}_j], \overline{H}_j], \overline{H}_j]\,.$$

By definition (22) and (36)

$$\begin{aligned} \mathcal{S}_j^{(3)} &= [\mathcal{S}_j^{(2)}, \widehat{H}_j] + \sigma_j\big(\mathcal{S}_j^{(2)}\big) \\ &= [\mathcal{S}_j^{(2)}, \widehat{H}_j] + 2\,\big(\mathcal{S}_j^{(1)} A_j^{[1]} + B_j^{[1]}\, \mathcal{S}_j^{(1)}\big) + \mathcal{S}_j^{(0)} A_j^{[2]} + B_j^{[2]}\, \mathcal{S}_j^{(0)}\,; \end{aligned}$$

due to $\mathcal{S}_j^{(0)}(0) = I$, $\mathcal{S}_j^{(1)}(0) = 0$, and $\mathcal{S}_j^{(2)}(0) = A_j^{[1]} + B_j^{[1]}$, evaluation at zero yields

$$\mathcal{S}_j^{(3)}(0) = [A_j^{[1]} + B_j^{[1]}, \widehat{H}_j] + A_j^{[2]} + B_j^{[2]} \,.$$

Thus, the integral representation

$$\begin{aligned}
\mathcal{S}_j^{(3)}(t) = \mathrm{e}^{tB_j} & \big([A_j^{[1]} + B_j^{[1]}, \widehat{H}_j] + A_j^{[2]} + B_j^{[2]} \big) \, \mathrm{e}^{tA_j} \\
& + 3 \int_0^t \mathrm{e}^{(t-\tau)B_j} \big(\mathcal{S}_j^{(2)}(\tau) \, A_j^{[1]} + B_j^{[1]} \, \mathcal{S}_j^{(2)}(\tau) \big) \, \mathrm{e}^{(t-\tau)A_j} \, \mathrm{d}\tau \\
& \quad + 3 \int_0^t \mathrm{e}^{(t-\tau)B_j} \big(\mathcal{S}_j^{(1)}(\tau) \, A_j^{[2]} + B_j^{[2]} \, \mathcal{S}_j^{(1)}(\tau) \big) \, \mathrm{e}^{(t-\tau)A_j} \, \mathrm{d}\tau \\
& \qquad + \int_0^t \mathrm{e}^{(t-\tau)B_j} \big(\mathcal{S}_j^{(0)}(\tau) \, A_j^{[3]} + B_j^{[3]} \, \mathcal{S}_j^{(0)}(\tau) \big) \, \mathrm{e}^{(t-\tau)A_j} \, \mathrm{d}\tau
\end{aligned} \tag{43}$$

follows, where $\mathcal{S}_j^{(1)}$ and $\mathcal{S}_j^{(2)}$ are expressed by (30) and (37), respectively. Analogously to the preceeding step, in order to ensure that the leading term in $\mathcal{S}^{(3)}$, given by

$$\begin{aligned}
\mathcal{S}^{(3;0)}(t) = \mathrm{e}^{tB_3} & \big([A_3^{[1]} + B_3^{[1]}, \widehat{H}_3] + A_3^{[2]} + B_3^{[2]} \big) \, \mathrm{e}^{tA_3} \, \mathcal{S}_2^{(0)}(t) \, \mathcal{S}_1^{(0)}(t) \\
& + \mathcal{S}_3^{(0)}(t) \, \mathrm{e}^{tB_2} \big([A_2^{[1]} + B_2^{[1]}, \widehat{H}_2] + A_2^{[2]} + B_2^{[2]} \big) \, \mathrm{e}^{tA_2} \, \mathcal{S}_1^{(0)}(t) \\
& \quad + \mathcal{S}_3^{(0)}(t) \, \mathcal{S}_2^{(0)}(t) \, \mathrm{e}^{tB_1} \big([A_1^{[1]} + B_1^{[1]}, \widehat{H}_1] + A_1^{[2]} + B_1^{[2]} \big) \, \mathrm{e}^{tA_1} \,,
\end{aligned}$$

satisfies $\mathcal{S}^{(3;0)}(t) \, v = \mathscr{O}(t)$, we employ the third-order conditions

$$\begin{aligned}
&\text{OC 3:} \qquad \mathcal{S}^{(3)}(0) = \sum_{j=1}^{s} \big([A_j^{[1]} + B_j^{[1]}, \widehat{H}_j] + A_j^{[2]} + B_j^{[2]} \big) = 0 \\
&\iff \quad \sum_{j=1}^{s} \Bigg(2 \sum_{k=1}^{j} \sum_{\ell=1}^{j} b_j \, a_k \, a_\ell - \sum_{j=1}^{s} \sum_{k=1}^{j-1} a_j \, b_k \, \Big(a_j + 2 \sum_{\ell=1}^{j-1} a_\ell \Big) \Bigg) = 0 \\
&\qquad \text{and} \quad \sum_{j=1}^{s} \Bigg(\sum_{k=1}^{j} \sum_{\ell=1}^{j} b_j \, a_k \, b_\ell - 2 \sum_{k=1}^{j-1} \sum_{\ell=1}^{j-1} a_j \, b_k \, b_\ell \Bigg) = 0 \,,
\end{aligned} \tag{44}$$

which ensure $\mathcal{S}^{(3;0)}(0) = 0$. By means of the variation-of-constants formula we obtain

$$\mathcal{S}^{(3;0)}(t) = \int_0^t \mathrm{e}^{(t-\tau)H} \, \delta\mathcal{S}^{(3;0)}(\tau) \, \mathrm{d}\tau \,,$$

and as desired $\mathcal{S}^{(3;0)}(t) = \mathscr{O}(t)$. Furthermore, the relation $\mathcal{S}^{(3)}(t) = \mathscr{O}(t)$ readily follows. In order to specify the structure of $\mathcal{S}^{(3;0)}$, arguments as in Section 4.3 are used, but with

$$\begin{aligned}\mathcal{X}_j(t) &= \mathrm{e}^{tB_j}\big([A_j^{[1]} + B_j^{[1]}, \widehat{H}_j] + A_j^{[2]} + B_j^{[2]}\big)\,\mathrm{e}^{tA_j}\,,\\ \mathcal{X}_j(0) &= [A_j^{[1]} + B_j^{[1]}, \widehat{H}_j] + A_j^{[2]} + B_j^{[2]}\,.\end{aligned}$$

Altogether, this shows that $\mathcal{S}^{(3)}$ can be expressed via compositions of certain iterated commutators and evolution operators.

Local error expansion. A local error expansion ensuring $\mathcal{L}(t)\,v = \mathscr{O}(t^4)$ under suitable regularity requirements on v is finally obtained by inserting the above integral representations for $\mathcal{S}_j^{(1)}, \mathcal{S}_j^{(2)}, \mathcal{S}_j^{(3)}$ into (40), see also (24a). In the context of linear Schrödinger equations involving sufficiently regular potentials, the evolution operators e^{tH}, e^{tA}, e^{tB} preserve the regularity properties of their arguments [10, Lemma 12]; thus it remains to deduce suitable bounds for iterated commutators, which leads to the regularity requirement $v \in H^3$, see Section 7. The main tools for a generalization to higher-order splitting methods are deduced in Section 6.

Proposition 1 (A priori local error expansion). *Provided that the considered three-stage exponential operator splitting method satisfies the third-order conditions* (32)*,* (38)*, and* (44)*, for the associated local error it follows*

$$\mathcal{L}(t)\,v = \mathscr{O}(t^4)\,,$$

under appropriate regularity requirements on the argument v. The local error expansion in particular comprises third iterated commutators of the involved operators A, B and the evolution operators e^{tH}*,* e^{tA}*,* e^{tB}*.*

4.5. Structure of the term $\mathcal{S}^{(4)}$

For $p = 3$ and in view of the analysis of our a posteriori local error estimator in Section 5 below, the structure of the quantity $\mathcal{S}^{(4)} = \delta\mathcal{S}^{(3)}$ is relevant. For a general scheme involving s stages, Lemma 3 shows

$$\mathcal{S}^{(4)} = \sum_{k_1+\cdots+k_s=4} \tfrac{4!}{k_1!\cdots k_s!}\, \mathcal{S}_s^{(k_s)} \cdots \mathcal{S}_1^{(k_1)}\,, \tag{45}$$

where it remains to specify the quantities $\mathcal{S}_j^{(4)}$. Invoking once more Lemma 4 with $k = 4$ yields

$$\begin{aligned}\sigma\big(\mathcal{S}_j^{(4)}\big) &= \tfrac{\mathrm{d}}{\mathrm{d}t}\mathcal{S}_j^{(4)} - \mathcal{S}_j^{(4)} A_j - B_j\, \mathcal{S}_j^{(4)} \\ &= 4\,\big(\mathcal{S}_j^{(3)} A_j^{[1]} + B_j^{[1]}\, \mathcal{S}_j^{(3)}\big) \\ &\quad + 6\,\big(\mathcal{S}_j^{(2)} A_j^{[2]} + B_j^{[2]}\, \mathcal{S}_j^{(2)}\big) \\ &\quad + 4\,\big(\mathcal{S}_j^{(1)} A_j^{[3]} + B_j^{[3]}\, \mathcal{S}_j^{(1)}\big) \\ &\quad + \mathcal{S}_j^{(0)} A_j^{[4]} + B_j^{[4]}\, \mathcal{S}_j^{(0)}\,,\end{aligned} \tag{46}$$

involving in particular the fourth iterated commutators

$$A_j^{[4]} = [[[[A_j, \overline{H}_{j-1}], \overline{H}_{j-1}], \overline{H}_{j-1}], \overline{H}_{j-1}]\,, \quad B_j^{[4]} = [[[[B_j, \overline{H}_j], \overline{H}_j], \overline{H}_j], \overline{H}_j]\,.$$

We thus obtain the integral representation

$$\begin{aligned}\mathcal{S}_j^{(4)}(t) &= \mathrm{e}^{tB_j}\, \mathcal{S}_j^{(4)}(0)\, \mathrm{e}^{tA_j} \\ &\quad + 4 \int_0^t \mathrm{e}^{(t-\tau)B_j}\, \big(\mathcal{S}_j^{(3)}(\tau)\, A_j^{[1]} + B_j^{[1]}\, \mathcal{S}_j^{(3)}(\tau)\big)\, \mathrm{e}^{(t-\tau)A_j}\, \mathrm{d}\tau \\ &\quad + 6 \int_0^t \mathrm{e}^{(t-\tau)B_j}\, \big(\mathcal{S}_j^{(2)}(\tau)\, A_j^{[2]} + B_j^{[2]}\, \mathcal{S}_j^{(2)}(\tau)\big)\, \mathrm{e}^{(t-\tau)A_j}\, \mathrm{d}\tau \\ &\quad + 4 \int_0^t \mathrm{e}^{(t-\tau)B_j}\, \big(\mathcal{S}_j^{(1)}(\tau)\, A_j^{[3]} + B_j^{[3]}\, \mathcal{S}_j^{(1)}(\tau)\big)\, \mathrm{e}^{(t-\tau)A_j}\, \mathrm{d}\tau \\ &\quad + \int_0^t \mathrm{e}^{(t-\tau)B_j}\, \big(\mathcal{S}_j^{(0)}(\tau)\, A_j^{[4]} + B_j^{[4]}\, \mathcal{S}_j^{(0)}(\tau)\big)\, \mathrm{e}^{(t-\tau)A_j}\, \mathrm{d}\tau\,,\end{aligned} \tag{47}$$

where

$$\begin{aligned}\mathcal{S}_j^{(4)}(0) &= [[A_j^{[1]} + B_j^{[1]}, \widehat{H}_j], \widehat{H}_j] + [A_j^{[2]} + B_j^{[2]}, \widehat{H}_j] \\ &\quad + 3\,\big((A_j^{[1]} + B_j^{[1]})\, A_j^{[1]} + B_j^{[1]}\, (A_j^{[1]} + B_j^{[1]})\big) + A_j^{[3]} + B_j^{[3]}\,,\end{aligned}$$

with $\mathcal{S}_j^{(4)}(0) \neq 0$, in general. In the context of Schrödinger equations this shows that the dominant terms in $\mathcal{S}^{(4)}$ involving fourth iterated commutators impose the regularity requirement $v \in H^4$ to ensure $\mathcal{S}^{(4)}(t) = \mathscr{O}(1)$. We note that the quantities $\mathcal{S}_j^{(4)}(0)$ contain compositions of iterated commutators, which remain bounded under the regularity requirements imposed on the leading terms.

Remark 1. Let us recapitulate the structure of the third-order conditions for a splitting method involving s stages. In addition to the basic consistency condition

$$\text{OC 1:} \qquad \mathcal{S}^{(1)}(0) = 0 \quad \Longleftrightarrow \quad \sum_{j=1}^{s} \big(A_j + B_j\big) = A + B\,,$$

it is required that the conditions

$$\begin{aligned}
&\text{OC 2:} \qquad \mathcal{S}^{(2)}(0) = \sum_{j=1}^{s} \big(A_j^{[1]} + B_j^{[1]}\big) = 0\,,\\
&\text{OC 3:} \qquad \mathcal{S}^{(3)}(0) = \sum_{j=1}^{s} \big([A_j^{[1]} + B_j^{[1]}, \widehat{H}_j] + A_j^{[2]} + B_j^{[2]}\big) = 0\,,
\end{aligned}$$

hold, see also (32), (38), and (44). We point out that $\mathcal{S}^{(2)}(0)$ is a multiple of the first commutator $[A, B]$, provided that the condition OC 1 is satisfied, and that $\mathcal{S}^{(3)}(0)$ is a linear combination of the second iterated commutators $[A, [A, B]]$ and $[B, [A, B]]$, provided that the conditions OC 1 and OC 2 are satisfied. However, if the validity of the respective lower-order conditions is not utilized, the quantities $\mathcal{S}^{(2)}(0)$ and $\mathcal{S}^{(3)}(0)$ are of a more complicated structure, involving terms that cannot be represented as commutators.

The above considerations also extend to high-order methods. For instance, a close inspection shows that the term

$$\begin{aligned}
\mathcal{S}^{(4)}(0) &= \mathcal{S}_3^{(4)}(0) + \mathcal{S}_2^{(4)}(0) + \mathcal{S}_1^{(4)}(0)\\
&\qquad + 6\,\big(\mathcal{S}_3^{(2)}(0)\,\mathcal{S}_2^{(2)}(0) + \mathcal{S}_3^{(2)}(0)\,\mathcal{S}_1^{(2)}(0) + \mathcal{S}_2^{(2)}(0)\,\mathcal{S}_1^{(2)}(0)\big)
\end{aligned} \tag{48}$$

reduces to a linear combination of the third iterated commutators $[A, [A, [A, B]]]$, $[B, [A, [A, B]]] = [A, [B, [A, B]]]$, and $[B, [B, [A, B]]]$ provided that the conditions OC 1, OC 2, and OC 3 are satisfied. The direct verification of this fact requires rather tedious calculations which we do not explicate here. It can be shown that such a structure of the order conditions is valid for splitting methods of arbitrary order p; however, a rigorous proof of this fact is beyond the scope of the present manuscript and will be given in a separate work. Such a result also implies that the order conditions obtained in this way are non-redundant. Furthermore, this enables the automatic generation of the respective system of polynomial equations for the coefficients $(a_j, b_j)_{1 \leq j \leq s}$ with the help of computer algebra.

5. A posteriori local error estimators

Construction of local error estimators ($s = p = 3$). As indicated before, for an exponential operator splitting method of the form (3) the construction of the defect-based local error estimator (7) relies on the application of an Hermite quadrature formula (6) for the approximation of (4d). In particular, for a three-stage third-order splitting method, application of the fourth-order Hermite quadrature formula

$$
\begin{aligned}
&\mathcal{Q}_f(t) = t\left(\tfrac{3}{4}\,f(0;t) + \tfrac{1}{4}\,t\,\partial_\tau f(0;t) + \tfrac{1}{24}\,t^2\,\partial_\tau^2 f(0;t) + \tfrac{1}{4}\,f(t;t)\right), \\
&\int_0^t f(\tau;t)\,\mathrm{d}\tau - \mathcal{Q}_f(t) = \mathscr{O}\big(t^5\big)\,, \quad t \geq 0\,,
\end{aligned}
\tag{49}
$$

see also (6), yields the local error estimator

$$
\begin{aligned}
\mathcal{P}(t) = \tfrac{1}{4}\,t\,\mathcal{S}^{(1)}(t) \quad &\approx \quad \mathcal{L}(t) = \int_0^t f(\tau;t)\,\mathrm{d}\tau\,, \\
f(\tau;t) &= \mathrm{e}^{(t-\tau)H}\,\mathcal{S}^{(1)}(\tau)\,, \quad 0 \leq \tau \leq t\,, \\
\mathcal{S}^{(1)} = \mathcal{S}_3^{(1)}\,\mathcal{S}_2^{(0)}\,\mathcal{S}_1^{(0)} &+ \mathcal{S}_3^{(0)}\,\mathcal{S}_2^{(1)}\,\mathcal{S}_1^{(0)} + \mathcal{S}_3^{(0)}\,\mathcal{S}_2^{(0)}\,\mathcal{S}_1^{(1)}\,,
\end{aligned}
\tag{50}
$$

see (4d), (24a), (27), and recall that the defect $\mathcal{D}$ equals $\mathcal{S}^{(1)}$. In fact, due to

$$
\begin{aligned}
f(\tau;t) &= \mathrm{e}^{(t-\tau)H}\,\mathcal{S}^{(1)}(\tau)\,, \\
\partial_\tau f(\tau;t) &= \mathrm{e}^{(t-\tau)H}\,\delta\mathcal{S}^{(1)}(\tau) = \mathrm{e}^{(t-\tau)H}\,\mathcal{S}^{(2)}(\tau)\,, \\
\partial_\tau^2 f(\tau;t) &= \mathrm{e}^{(t-\tau)H}\,\delta\mathcal{S}^{(2)}(\tau) = \mathrm{e}^{(t-\tau)H}\,\mathcal{S}^{(3)}(\tau)\,,
\end{aligned}
\tag{51}
$$

and the validity of the conditions $\mathcal{S}^{(1)}(0) = \mathcal{S}^{(2)}(0) = \mathcal{S}^{(3)}(0) = 0$ reflecting the third-order conditions it follows

$$
f(0;t) = \partial_\tau f(0;t) = \partial_\tau^2 f(0;t) = 0\,, \qquad f(t;t) = \mathcal{S}^{(1)}(t)\,,
$$

hence $\mathcal{P}(t) = \frac{1}{4}\,t\,\mathcal{S}^{(1)}(t)$ results, see (50).

Construction of local error estimators (general case). More generally, following the analogous approach, for a p-th-order splitting method the defect-based local error estimator is given by

$$
\mathcal{P}(t) = \tfrac{1}{p+1}\,t\,\mathcal{S}^{(1)}(t) \approx \mathcal{L}(t)\,,
$$

which leads to the representation (7).

Asymptotical correctness ($s = p = 3$). The analysis comprises three steps.

(i) For the Hermite quadrature formula (49) the Peano representation for the quadrature error involving a third-order Peano kernel reads

$$\mathcal{P}(t) - \mathcal{L}(t) = \int_0^t K_3(\tau;t)\, \partial_\tau^3 f(\tau;t)\, \mathrm{d}\tau\,, \quad K_3(\tau;t) = \tfrac{1}{24}\,(4\tau - t)\,(t-\tau)^2\,,$$

where $K_3(\tau;t)$ satisfies

$$\int_0^t K_3(\tau;t)\, \mathrm{d}\tau = 0\,. \tag{52}$$

Further differentiation of (51) implies $\partial_\tau^3 f(\tau;t) = \mathrm{e}^{(t-\tau)H}\, \mathcal{S}^{(4)}(\tau)$; this yields the representation

$$\mathcal{P}(t) - \mathcal{L}(t) = \int_0^t K_3(\tau;t)\, \mathrm{e}^{(t-\tau)H}\, \mathcal{S}^{(4)}(\tau)\, \mathrm{d}\tau\,.$$

Our basic idea is to exploit relation (52) and to rewrite $\mathcal{P} - \mathcal{L}$ as

$$\begin{aligned}\mathcal{P}(t) - \mathcal{L}(t) &= \mathrm{e}^{tH}\, \mathcal{S}^{(4)}(0) \int_0^t K_3(\tau;t)\, \mathrm{d}\tau \\ &\qquad + \int_0^t K_3(\tau;t)\, \mathrm{e}^{(t-\tau)H} \big(\mathcal{S}^{(4)}(\tau) - \mathrm{e}^{\tau H} \mathcal{S}^{(4)}(0)\big)\, \mathrm{d}\tau \\ &= \int_0^t K_3(\tau;t)\, \mathrm{e}^{(t-\tau)H} \big(\mathcal{S}^{(4)}(\tau) - \mathrm{e}^{\tau H} \mathcal{S}^{(4)}(0)\big)\, \mathrm{d}\tau\,.\end{aligned}$$

Evidently, the relation $K_3(\tau;t) = \mathscr{O}(t^3)$ holds; consequently, in order to ensure $\mathcal{P}(t) - \mathcal{L}(t) = \mathscr{O}(t^5)$, a close inspection of the leading term in $\mathrm{e}^{(t-\tau)H}\big(\mathcal{S}^{(4)}(\tau) - \mathrm{e}^{\tau H}\mathcal{S}^{(4)}(0)\big)$ is required.

(ii) Recall the structure of $\mathcal{S}^{(4)}(0)$ specified in (48), and consider the time evolution of the corresponding operator

$$\begin{aligned}&\mathcal{S}_3^{(4)}\mathcal{S}_2^{(0)}\mathcal{S}_1^{(0)} + \mathcal{S}_3^{(0)}\mathcal{S}_2^{(4)}\mathcal{S}_1^{(0)} + \mathcal{S}_3^{(0)}\mathcal{S}_2^{(0)}\mathcal{S}_1^{(4)} \\ &\quad + 6\big(\mathcal{S}_3^{(2)}\mathcal{S}_2^{(2)}\mathcal{S}_1^{(0)} + \mathcal{S}_3^{(2)}\mathcal{S}_2^{(0)}\mathcal{S}_1^{(2)} + \mathcal{S}_3^{(0)}\mathcal{S}_2^{(2)}\mathcal{S}_1^{(2)}\big).\end{aligned}$$

In this expression, replace all $\mathcal{S}_j^{(k)}$ by

$$\tilde{\mathcal{S}}_j^{(k)}(t) := \mathrm{e}^{tB_j}\, \mathcal{S}_j^{(k)}(0\,)\, \mathrm{e}^{tA_j}, \qquad \tilde{\mathcal{S}}_j^{(k)}(0) = \mathcal{S}_j^{(k)}(0),$$

and consider $\tilde{\mathcal{S}}^{(4)}$, defined by

$$\begin{aligned}\tilde{\mathcal{S}}^{(4)} := \tilde{\mathcal{S}}_3^{(4)}\tilde{\mathcal{S}}_2^{(0)}\tilde{\mathcal{S}}_1^{(0)} + \tilde{\mathcal{S}}_3^{(0)}\tilde{\mathcal{S}}_2^{(4)}\tilde{\mathcal{S}}_1^{(0)} + \tilde{\mathcal{S}}_3^{(0)}\tilde{\mathcal{S}}_2^{(0)}\tilde{\mathcal{S}}_1^{(4)} \\ + 6\,\big(\tilde{\mathcal{S}}_3^{(2)}\tilde{\mathcal{S}}_2^{(2)}\tilde{\mathcal{S}}_1^{(0)} + \tilde{\mathcal{S}}_3^{(2)}\tilde{\mathcal{S}}_2^{(0)}\tilde{\mathcal{S}}_1^{(2)} + \tilde{\mathcal{S}}_3^{(0)}\tilde{\mathcal{S}}_2^{(2)}\tilde{\mathcal{S}}_1^{(2)}\big).\end{aligned}$$

Here,

$$\tilde{\mathcal{S}}_3^{(k_3)}(t)\,\tilde{\mathcal{S}}_2^{(k_2)}(t)\,\tilde{\mathcal{S}}_1^{(k_1)}(t) \tag{53a}$$

are splitting analogues of

$$\mathrm{e}^{tH}\,\mathcal{S}_3^{(k_3)}(0)\,\mathcal{S}_2^{(k_2)}(0)\,\mathcal{S}_1^{(k_1)}(0). \tag{53b}$$

To study the difference between (53a) and (53b), we apply again (21) and insert $\sigma_j(\tilde{\mathcal{S}}_j^{(k_j)}) = 0$, obtaining

$$\begin{aligned}\delta\big(\tilde{\mathcal{S}}_3^{(k_3)}\,\tilde{\mathcal{S}}_2^{(k_2)}\,\tilde{\mathcal{S}}_1^{(k_1)}\big) = [\tilde{\mathcal{S}}_3^{(k_3)}, \widehat{H}_3]\,\tilde{\mathcal{S}}_2^{(k_2)}\,\tilde{\mathcal{S}}_1^{(k_1)} \\ + \tilde{\mathcal{S}}_3^{(k_3)}\,[\tilde{\mathcal{S}}_2^{(k_2)}, \widehat{H}_2]\,\tilde{\mathcal{S}}_1^{(k_1)} \\ + \tilde{\mathcal{S}}_3^{(k_3)}\,\tilde{\mathcal{S}}_2^{(k_2)}\,[\tilde{\mathcal{S}}_1^{(k_1)}, \widehat{H}_1]\,.\end{aligned}$$

Again, we represent $[\tilde{\mathcal{S}}_j^{(k_j)}, \widehat{H}_j]$ as solutions of Sylvester equations, (see Lemma 1, (iii)). This results in an evolution equation of the form

$$\delta\big(\tilde{\mathcal{S}}_3^{(k_3)}(t)\,\tilde{\mathcal{S}}_2^{(k_2)}(t)\,\tilde{\mathcal{S}}_1^{(k_1)}(t) - \mathrm{e}^{tH}\,\mathcal{S}_3^{(k_3)}(0)\,\mathcal{S}_2^{(k_2)}(0)\,\mathcal{S}_1^{(k_1)}(0)\big) = \mathscr{O}(1), \tag{54}$$

with homogeneous initial condition. We note that the inhomogeneity in (54) is $\mathscr{O}(1)$ but not $\mathscr{O}(t)$, which is sufficient in the present context. This is due to the fact that for $k_j > 0$ the initial values $[\tilde{\mathcal{S}}_j^{(k_j)}(0), \widehat{H}_j] = [\mathcal{S}_j^{(k_j)}(0), \widehat{H}_j]$ are commutator expressions which do not vanish, in contrast to $[\mathcal{S}_j^{(0)}(0), \widehat{H}_j] = [I, \widehat{H}_j] = 0$.

Summing up all the contributions, after integration we obtain

$$\tilde{\mathcal{S}}^{(4)}(t) - \mathrm{e}^{tH}\,\mathcal{S}^{(4)}(0) = \mathscr{O}(t).$$

(iii) Noting that

$$\mathrm{e}^{\tau H}\,S^{(4)}(0) - S^{(4)}(t) = \mathrm{e}^{\tau H}\,S^{(4)}(0) - \tilde{\mathcal{S}}^{(4)}(t) + \tilde{\mathcal{S}}^{(4)}(t) - S^{(4)}(t)\,,$$

it remains to study the second term $\tilde{\mathcal{S}}^{(4)}(t) - S^{(4)}(t)$, which consists of

- $\mathscr{O}(t)$-terms in the multinomial expansion (45) for $S^{(4)}(t)$ (comprising all other index combinations as for instance $(2,1,1)$ with homogeneous initial value), and
- terms like, for instance,

$$\tilde{\mathcal{S}}_3^{(2)}\,\tilde{\mathcal{S}}_2^{(2)}\,\tilde{\mathcal{S}}_1^{(0)} - \mathcal{S}_3^{(2)}\,\mathcal{S}_2^{(2)}\,\mathcal{S}_1^{(0)}$$

  wish vanish at $t = 0$.

For the latter terms, $\mathscr{O}(t)$ remains to be shown. After rearranging, the triangle inequality implies that it is sufficient to verify

$$\tilde{\mathcal{S}}_j^{(k)}(t) - \mathcal{S}_j^{(k)}(t) = \mathscr{O}(t).$$

These terms are given by the integral representations for $\mathcal{S}_j^{(k)}(t)$ where the $\mathscr{O}(1)$ terms $\tilde{\mathcal{S}}_j^{(k)}(t) = \mathrm{e}^{tB_j}\,\mathcal{S}_j^{(k)}(0)\,\mathrm{e}^{tA_j}$ cancel out, cf. for example (47) for $k = 4$.

Altogether, this shows asymptotical correctness of the local error estimator $\mathcal{P}(t)$ from (50). $\square$

Proposition 2 (Asymptotical correctness). *Provided that the considered three-stage exponential operator splitting method satisfies the third-order conditions* (32), (38), *and* (44), *the associated defect-based local error estimator is asymptotically correct, that is, it holds*

$$\big(\mathcal{P}(t) - \mathcal{L}(t)\big)\,v = \mathscr{O}\big(t^5\big)$$

under appropriate regularity requirements on the argument v. The above expansion in particular comprises fourth iterated commutators of the involved operators A, B and the evolution operators e^{tH}, e^{tA}, e^{tB}.

Asymptotical correctness (General case). More generally, for a p-th-order splitting method we obtain the following Peano representation for the quadrature approximation error

$$\begin{aligned}\mathcal{P}(t) - \mathcal{L}(t) &= \int_0^t K_p(\tau;t)\,\partial_\tau^p\,f(\tau;t)\,\mathrm{d}\tau\\ &= \int_0^t K_p(\tau;t)\,\mathrm{e}^{(t-\tau)H}\,\mathcal{S}^{(p+1)}(\tau)\ \mathrm{d}\tau\,;\end{aligned}$$

here, the p-th order Peano kernel K_p is a polynomial of degree p in τ satisfying

$$\int_0^t K_p(\tau;t)\,\mathrm{d}\tau = 0\,.$$

Rewriting the difference $\mathcal{P}-\mathcal{L}$ in a similar manner as before shows that the a posteriori local error estimator quadrature approximation is asymptotically correct,

$$\mathcal{P}(t)\,v - \mathcal{L}(t)\,v = \mathscr{O}\big(t^{p+2}\big)\,,$$

provided that the argument v satisfies suitable regularity requirements. In the context of linear Schrödinger equations the neccessary regularity assumptions are specified in Section 7; auxiliary results on the structure of $\mathcal{S}^{(p+1)}$ are provided in Section 6 below.

6. Main tools for a generalization to higher-order splitting methods

In the following, we derive auxiliary results specifying the structure of the operator $\mathcal{S}^{(n)}$ for arbitrary $n \geq 1$; as indicated in Sections 4 and 5 these results provide the main ingredients for an extension of our approach to higher-order exponential operator splitting methods (3).

Evolution equation for multiple product. The following auxiliary result provides a relation for the derivative of a multiple product of time-dependent operators $\mathcal{X}_\nu$ satisfying a Sylvester equation. We denote

$$\mathcal{X}_\ell^k = \prod_{\nu=\ell}^{k} \mathcal{X}_\nu = \mathcal{X}_k\,\mathcal{X}_{k-1}\,\cdots\,\mathcal{X}_\ell\,, \quad k \geq \ell\,, \qquad \mathcal{X}_\ell^k = I\,, \quad k < \ell\,. \tag{55}$$

Lemma 2. *For any $1 \leq j \leq s$ the product $\mathcal{X}_1^j = \mathcal{X}_j \cdots \mathcal{X}_1$ of time-dependent operators satisfying the inhomogeneous Sylvester equations*

$$\begin{cases} \frac{\mathrm{d}}{\mathrm{d}t}\mathcal{X}_\nu = \mathcal{X}_\nu\,A_\nu + B_\nu\,\mathcal{X}_\nu + \mathcal{G}_\nu\,, \\ \mathcal{X}_\nu(0)\ \textit{given}\,, \end{cases} \tag{56}$$

is a solution of the initial value problem

$$\begin{cases} \frac{\mathrm{d}}{\mathrm{d}t}\mathcal{X}_1^j = \overline{H}_j\,\mathcal{X}_1^j + \displaystyle\sum_{\nu=1}^{j} \mathcal{X}_{\nu+1}^j\,\big([\mathcal{X}_\nu,\widehat{H}_\nu] + \mathcal{G}_\nu\big)\,\mathcal{X}_1^{\nu-1}\,, \\ \mathcal{X}_1^j(0)\ \textit{given.} \end{cases} \tag{57a}$$

In particular, the operator $\mathcal{X} = \mathcal{X}_1^s = \mathcal{X}_s \cdots \mathcal{X}_1$ *satisfies*

$$\begin{cases} \frac{\mathrm{d}}{\mathrm{d}t}\mathcal{X} = H\,\mathcal{X} + \sum_{\nu=1}^{s} \mathcal{X}_{\nu+1}^{s}\big([\mathcal{X}_\nu, \widehat{H}_\nu] + \mathcal{G}_\nu\big)\,\mathcal{X}_1^{\nu-1}\,, \\ \mathcal{X}(0) \text{ given.} \end{cases} \tag{57b}$$

PROOF. We apply induction on j.

- For $j=1$ assertion (57a) follows at once from (56), since

$$\begin{aligned} \tfrac{\mathrm{d}}{\mathrm{d}t}\mathcal{X}_1 &= \mathcal{X}_1\,A_1 + B_1\,\mathcal{X}_1 + \mathcal{G}_1 \\ &= (A_1 + B_1)\,\mathcal{X}_1 + [\mathcal{X}_1, A_1] + \mathcal{G}_1 \\ &= H_1\,\mathcal{X}_1 + [\mathcal{X}_1, \widehat{H}_1] + \mathcal{G}_1\,. \end{aligned}$$

- In order to prove the induction step $j-1 \to j$ for $2 \le j \le s$, we make use of the commutator identity (10). Differentiation and application of the induction assumption yields

$$\begin{aligned} \tfrac{\mathrm{d}}{\mathrm{d}t}\mathcal{X}_1^j &= \tfrac{\mathrm{d}}{\mathrm{d}t}\big(\mathcal{X}_j\,\mathcal{X}_1^{j-1}\big) = \tfrac{\mathrm{d}}{\mathrm{d}t}(\mathcal{X}_j)\,\mathcal{X}_1^{j-1} + \mathcal{X}_j\,\tfrac{\mathrm{d}}{\mathrm{d}t}\mathcal{X}_1^{j-1} \\ &= \big(\mathcal{X}_j\,A_j + B_j\,\mathcal{X}_j + \mathcal{G}_j\big)\,\mathcal{X}_1^{j-1} \\ &\quad + \mathcal{X}_j\left(\overline{H}_{j-1}\,\mathcal{X}_1^{j-1} + \sum_{\nu=1}^{j-1}\mathcal{X}_{\nu+1}^{j-1}\big([\mathcal{X}_\nu, \widehat{H}_\nu] + \mathcal{G}_\nu\big)\,\mathcal{X}_1^{\nu-1}\right) \\ &= \big(H_j\,\mathcal{X}_j + [\mathcal{X}_j, A_j] + \mathcal{G}_j\big)\,\mathcal{X}_1^{j-1} \\ &\quad + \mathcal{X}_j\,\overline{H}_{j-1}\,\mathcal{X}_1^{j-1} + \sum_{\nu=1}^{j-1}\mathcal{X}_{\nu+1}^{j}\big([\mathcal{X}_\nu, \widehat{H}_\nu] + \mathcal{G}_\nu\big)\,\mathcal{X}_1^{\nu-1} \\ &= H_j\,\mathcal{X}_1^j + \mathcal{X}_j\,\overline{H}_{j-1}\,\mathcal{X}_1^{j-1} + \big([\mathcal{X}_j, A_j] + \mathcal{G}_j\big)\,\mathcal{X}_1^{j-1} \\ &\quad + \sum_{\nu=1}^{j-1}\mathcal{X}_{\nu+1}^{j}\big([\mathcal{X}_\nu, \widehat{H}_\nu] + \mathcal{G}_\nu\big)\,\mathcal{X}_1^{\nu-1}\,. \end{aligned}$$

  Rearranging the first three terms according to

$$\begin{aligned} &H_j\,\mathcal{X}_1^j + \mathcal{X}_j\,\overline{H}_{j-1}\,\mathcal{X}_1^{j-1} + \big([\mathcal{X}_j, A_j] + \mathcal{G}_j\big)\,\mathcal{X}_1^{j-1} \\ &= H_j\,\mathcal{X}_1^j + \overline{H}_{j-1}\,\mathcal{X}_j\,\mathcal{X}_1^{j-1} + [\mathcal{X}_j, \overline{H}_{j-1}]\,\mathcal{X}_1^{j-1} + \big([\mathcal{X}_j, A_j] + \mathcal{G}_j\big)\,\mathcal{X}_1^{j-1} \\ &= H_j\,\mathcal{X}_1^j + \overline{H}_{j-1}\,\mathcal{X}_1^j + \big([\mathcal{X}_j, A_j + \overline{H}_{j-1}] + \mathcal{G}_j\big)\,\mathcal{X}_1^{j-1} \\ &= \overline{H}_j\,\mathcal{X}_1^j + \big([\mathcal{X}_j, \widehat{H}_j] + \mathcal{G}_j\big)\,\mathcal{X}_1^{j-1}\,, \end{aligned}$$

further implies

$$\begin{aligned}
\tfrac{\mathrm{d}}{\mathrm{d}t}\mathcal{X}_1^j &= \overline{H}_j\,\mathcal{X}_1^j + \big([\mathcal{X}_j,\widehat{H}_j] + \mathcal{G}_j\big)\,\mathcal{X}_1^{j-1} + \sum_{\nu=1}^{j-1} \mathcal{X}_{\nu+1}^j\,\big([\mathcal{X}_\nu,\widehat{H}_\nu] + \mathcal{G}_\nu\big)\,\mathcal{X}_1^{\nu-1} \\
&= \overline{H}_j\,\mathcal{X}_1^j + \sum_{\nu=1}^{j} \mathcal{X}_{\nu+1}^j\,\big([\mathcal{X}_\nu,\widehat{H}_\nu] + \mathcal{G}_\nu\big)\,\mathcal{X}_1^{\nu-1}\,,
\end{aligned}$$

which completes the induction argument. □

Representation for $\mathcal{S}^{(n)}$. In order to establish a local error expansion of the form (25) and to prove asymptotical correctness of the a posteriori local error estimator (7), it is essential to employ a suitable representation for $\mathcal{S}^{(n)} = \delta^n \mathcal{S}^{(0)}$.

Lemma 3. *The quantity $\mathcal{S}^{(n)}$ can be represented in the form*

$$\mathcal{S}^{(n)} = \sum_{k_1+\cdots+k_s=n} \tfrac{n!}{k_1!\cdots k_s!}\,\mathcal{S}_s^{(k_s)}\cdots\mathcal{S}_1^{(k_1)}\,, \tag{58a}$$

where $\mathcal{S}_j^{(k)}$ are recursively defined by

$$\mathcal{S}_j^{(0)} = \mathcal{S}_j\,, \qquad \mathcal{S}_j^{(k)} = [\mathcal{S}_j^{(k-1)},\widehat{H}_j] + \sigma_j\big(\mathcal{S}_j^{(k-1)}\big)\,, \quad k \geq 1\,. \tag{58b}$$

Proof. We refer to Sections 4 and 5 for a detailed treatment of the special case $s = p = 3$ and recall the notations (20) as well as (22). With the help of Lemma 2 providing the starting point for an induction argument

$$\mathcal{S}^{(1)} = \delta\mathcal{S}^{(0)} = \tfrac{\mathrm{d}}{\mathrm{d}t}\mathcal{S}^{(0)} - H\,\mathcal{S}^{(0)} = \sum_{\nu=1}^{s} \mathcal{S}_s^{(0)}\cdots\mathcal{S}_{\nu+1}^{(0)}\,[\mathcal{S}_\nu^{(0)},\widehat{H}_\nu]\,\mathcal{S}_{\nu-1}^{(0)}\cdots\mathcal{S}_1^{(0)}\,,$$

the proof of Lemma 3 is then identical with the proof of the general multinomial Leibniz formula for higher derivatives of a product of functions. In the present situation, the linear operation $\mathcal{S}_j^{(k-1)} \mapsto \mathcal{S}_j^{(k)}$ according to (58b) replaces the linear operation of differentiation. □

Representation of $\mathcal{S}_j^{(k)}$. In order to deduce an appropriate representation for $\mathcal{S}^{(n)}$ and $\mathcal{S}^{(n)}(t) - \mathrm{e}^{tH}\mathcal{S}^{(n)}(0)$, respectively, it is essential to represent the building blocks $\mathcal{S}_j^{(k)}$ in a suitable manner. This is accomplished by the following auxiliary result, which enables a representation of $\mathcal{S}_j^{(k)}$ as solution of a Sylvester-type equation with inhomogeneity depending on $\mathcal{S}_j^{(0)}, \dots, \mathcal{S}_j^{(k-1)}$.

Lemma 4. *The binomial expansion*

$$\tfrac{\mathrm{d}}{\mathrm{d}t}\mathcal{S}_j^{(k)} = \sum_{\ell=0}^{k} \binom{k}{\ell} \big(\mathcal{S}_j^{(\ell)} A_j^{[k-\ell]} + B_j^{[k-\ell]}\, \mathcal{S}_j^{(\ell)}\big) \tag{59a}$$

holds true for any $1 \leq j \leq s$ and $k \geq 0$; that is, $\mathcal{S}_j^{(k)}$ satisfies a Sylvester-type equation with inhomogeneity

$$\begin{aligned} \sigma_j(\mathcal{S}_j^{(k)}) &= \tfrac{\mathrm{d}}{\mathrm{d}t}\mathcal{S}_j^{(k)} - \mathcal{S}_j^{(k)} A_j - B_j\, \mathcal{S}_j^{(k)} \\ &= \sum_{\ell=0}^{k-1} \binom{k}{\ell} \big(\mathcal{S}_j^{(\ell)} A_j^{[k-\ell]} + B_j^{[k-\ell]}\, \mathcal{S}_j^{(\ell)}\big)\,. \end{aligned} \tag{59b}$$

Proof. For notational simplicity, we meanwhile suppress the stage index j. For given $A = A^{[0]}, B = B^{[0]}$ and $\widehat{H}$, define

$$\underline{H} = \widehat{H} - A\,, \quad \overline{H} = \widehat{H} + B\,,$$

(see (9a)), and

$$A^{[\ell]} = [A^{[\ell-1]}, \underline{H}]\,, \quad B^{[\ell]} = [B^{[\ell-1]}, \overline{H}]\,, \quad \ell \geq 1\,,$$

(see (11)). As in (20), we use the abbreviation

$$\sigma(\mathcal{X}) = \tfrac{\mathrm{d}}{\mathrm{d}t}\mathcal{X} - \mathcal{X} A - B\, \mathcal{X}\,.$$

Assume that $\mathcal{X}^{(0)}$ is given, satisfying $\sigma(\mathcal{X}^{(0)}) = 0$, and consider the recursion (see (58b))

$$\mathcal{X}^{(k)} = [\mathcal{X}^{(k-1)}, \widehat{H}] + \sigma(\mathcal{X}^{(k-1)})\,, \quad k \geq 1\,.$$

Now we prove the analogue to (59b),

$$\sigma(\mathcal{X}^{(k)}) = \sum_{\ell=0}^{k-1} \binom{k}{\ell} \big(\mathcal{X}^{(\ell)} A^{[k-\ell]} + B^{[k-\ell]}\, \mathcal{X}^{(\ell)}\big) \tag{60}$$

for all $k \geq 0$. In the following we repeatedly make use of identity (10).

- *Step 1:* For $\mu \geq 0$ the auxiliary identity
$$\begin{aligned}
&[\mathcal{X}A^{[\mu]} + B^{[\mu]}\mathcal{X}, \widehat{H}] + \sigma(\mathcal{X}A^{[\mu]} + B^{[\mu]}\mathcal{X}) \\
&= \mathcal{X}A^{[\mu+1]} + B^{[\mu+1]}\mathcal{X} \\
&\quad + \big([\mathcal{X}, \widehat{H}] + \sigma(\mathcal{X})\big)A^{[\mu]} + B^{[\mu]}\big([\mathcal{X}, \widehat{H}] + \sigma(\mathcal{X})\big)
\end{aligned} \tag{61}$$
holds, since
$$\begin{aligned}
&[\mathcal{X}A^{[\mu]} + B^{[\mu]}\mathcal{X}, \widehat{H}] + \sigma(\mathcal{X}A^{[\mu]} + B^{[\mu]}\mathcal{X}) \\
&= \mathcal{X}\,[A^{[\mu]}, \widehat{H}] + [\mathcal{X}, \widehat{H}]\,A^{[\mu]} + B^{[\mu]}\,[\mathcal{X}, \widehat{H}] + [B^{[\mu]}, \widehat{H}]\,\mathcal{X} \\
&\quad + \tfrac{\mathrm{d}}{\mathrm{d}t}\mathcal{X}A^{[\mu]} + B^{[\mu]}\tfrac{\mathrm{d}}{\mathrm{d}t}\mathcal{X} - \big(\mathcal{X}A^{[\mu]} + B^{[\mu]}\,\mathcal{X}\big)\,A - B\,\big(\mathcal{X}A^{[\mu]} + B^{[\mu]}\mathcal{X}\big) \\
&= \mathcal{X}\,[A^{[\mu]}, \widehat{H}] + [\mathcal{X}, \widehat{H}]\,A^{[\mu]} + B^{[\mu]}\,[\mathcal{X}, \widehat{H}] + [B^{[\mu]}, \widehat{H}]\,\mathcal{X} \\
&\quad + \big(\sigma(\mathcal{X}) + \mathcal{X}A + B\,\mathcal{X}\big)\,A^{[\mu]} + B^{[\mu]}\,\big(\sigma(\mathcal{X}) + \mathcal{X}A + B\,\mathcal{X}\big) \\
&\quad - \big(\mathcal{X}A^{[\mu]} + B^{[\mu]}\mathcal{X}\big)\,A - B\,\big(\mathcal{X}A^{[\mu]} + B^{[\mu]}\mathcal{X}\big) \\
&= \mathcal{X}\,[A^{[\mu]}, \widehat{H}] - \mathcal{X}\,[A^{[\mu]}, A] + [B^{[\mu]}, \widehat{H}]\,\mathcal{X} + [B^{[\mu]}, B]\,\mathcal{X} \\
&\quad + [\mathcal{X}, \widehat{H}]\,A^{[\mu]} + \sigma(\mathcal{X})\,A^{[\mu]} + B^{[\mu]}\,[\mathcal{X}, \widehat{H}] + B^{[\mu]}\,\sigma(\mathcal{X}) \\
&= \mathcal{X}A^{[\mu+1]} + B^{[\mu+1]}\mathcal{X} + \big([\mathcal{X}, \widehat{H}] + \sigma(\mathcal{X})\big)A^{[\mu]} + B^{[\mu]}\big([\mathcal{X}, \widehat{H}] + \sigma(\mathcal{X})\big)
\end{aligned}$$
after rearrangement, observing the recursive definition of $A^{[\ell]}$ and $B^{[\ell]}$.

- *Step 2:* For $k = 0$, (60) is equivalent to the assumption $\sigma(\mathcal{X}^{(0)}) = 0$.

- *Step 3:* Induction $k \to k+1$:
$$\begin{aligned}
\sigma(\mathcal{X}^{(k+1)}) &= \sigma([\mathcal{X}^{(k)}, \widehat{H}]) + \sigma(\sigma(\mathcal{X}^{[k]})) \\
&= \tfrac{\mathrm{d}}{\mathrm{d}t}[\mathcal{X}^{(k)}, \widehat{H}] - [\mathcal{X}^{(k)}, \widehat{H}]\,A - B\,[\mathcal{X}^{(k)}, \widehat{H}] + \sigma(\sigma(\mathcal{X}^{[k]})) \\
&= [\mathcal{X}^{(k)}\,A + B\,\mathcal{X}^{(k)}, \widehat{H}] - [\mathcal{X}^{(k)}, \widehat{H}]\,A - B\,[\mathcal{X}^{(k)}, \widehat{H}] \\
&\quad + [\sigma(\mathcal{X}^{(k)}), \widehat{H}] + \sigma(\sigma(\mathcal{X}^{[k]})) \\
&= \mathcal{X}^{(k)}\,[A, \widehat{H}] + [B, \widehat{H}]\,\mathcal{X}^{(k)} + [\sigma(\mathcal{X}^{(k)}), \widehat{H}] + \sigma(\sigma(\mathcal{X}^{[k]})) \\
&= \mathcal{X}^{(k)}[A, \underline{H}] + [B, \overline{H}]\,\mathcal{X}^{(k)} + [\sigma(\mathcal{X}^{(k)}), \widehat{H}] + \sigma(\sigma(\mathcal{X}^{[k]})) \\
&\overset{\text{ind}}{=} \mathcal{X}^{(k)}A^{[1]} + B^{[1]}\,\mathcal{X}^{(k)} \\
&\quad + \sum_{\ell=0}^{k-1} \binom{k}{\ell}\big(\,[\mathcal{X}^{(\ell)}A^{[k-\ell]} + B^{[k-\ell]}\,\mathcal{X}^{(\ell)}, \widehat{H}] \\
&\qquad\qquad + \sigma(\mathcal{X}^{(\ell)}A^{[k-\ell]} + B^{[k-\ell]}\,\mathcal{X}^{(\ell)})\big)\,.
\end{aligned}$$

Now, identity (61) together with the recursive definition of $\mathcal{X}^{(\ell)}$ implies

$$[\mathcal{X}^{(\ell)} A^{[k-\ell]} + B^{[k-\ell]}\, \mathcal{X}^{(\ell)}, \widehat{H}] + \sigma(\mathcal{X}^{(\ell)} A^{[k-\ell]} + B^{[k-\ell]}\, \mathcal{X}^{(\ell)})$$
$$= \mathcal{X}^{(\ell)} A^{[k+1-\ell]} + B^{[k+1-\ell]}\, \mathcal{X}^{(\ell)} + \big(\mathcal{X}^{(\ell+1)} A^{[k-\ell]} + B^{[k-\ell]}\, \mathcal{X}^{(\ell+1)}\big).$$

Thus, $\sigma(\mathcal{X}^{(k+1)})$ evaluates to

$$\begin{aligned}
\sigma(\mathcal{X}^{(k+1)}) &= \mathcal{X}^{(k)} A^{[1]} + B^{[1]}\, \mathcal{X}^{(k)} \\
&\quad + \sum_{\ell=0}^{k-1} \binom{k}{\ell} \big(\mathcal{X}^{(\ell)} A^{[k+1-\ell]} + B^{[k+1-\ell]}\, \mathcal{X}^{(\ell)}\big) \\
&\quad + \sum_{\ell=0}^{k-1} \binom{k}{\ell} \big(\mathcal{X}^{(\ell+1)} A^{[k-\ell]} + B^{[k-\ell]}\, \mathcal{X}^{(\ell+1)}\big) \\
&= \sum_{\ell=0}^{k} \binom{k}{\ell} \big(\mathcal{X}^{(\ell)} A^{[k+1-\ell]} + B^{[k+1-\ell]}\, \mathcal{X}^{(\ell)}\big) \\
&\quad + \sum_{\ell=1}^{k} \binom{k}{\ell-1} \big(\mathcal{X}^{(\ell)} A^{[k+1-\ell]} + B^{[k+1-\ell]}\, \mathcal{X}^{(\ell)}\big) \\
&= \mathcal{X}^{(0)} A^{[k+1]} + B^{[k+1]}\, \mathcal{X}^{(0)} \\
&\quad + \sum_{\ell=1}^{k} \binom{k+1}{\ell} \big(\mathcal{X}^{(\ell)} A^{[k+1-\ell]} + B^{[k+1-\ell]}\, \mathcal{X}^{(\ell)}\big) \\
&= \sum_{\ell=0}^{k} \binom{k+1}{\ell} \big(\mathcal{X}^{(\ell)} A^{[k+1-\ell]} + B^{[k+1-\ell]}\, \mathcal{X}^{(\ell)}\big)\,,
\end{aligned}$$

which proves (60).

Finally, setting $\mathcal{X}^{(k)} = \mathcal{S}_j^{(k)}$, the proposed identity (59) corresponds to (60) . $\square$

7. Application to linear Schrödinger equations

In this section, we discuss the application of our a priori and a posteriori local error analysis to linear Schrödinger equations (1); in particular, we state the regularity assumptions on the exact solution $\psi : \mathbb{R}^d \times [0,T] \to \mathbb{C}$ and the potential $V : \mathbb{R}^d \to \mathbb{R}$.

Main tools for the local error analysis. In our previous work [10] concerned with a posteriori local error estimators for (1) based on the first-order Lie–Trotter splitting method and the second-order Strang splitting method, respectively, it has been demonstrated that the obtained a priori and a posteriori local error expansions involve commutators of the operators $A = \mathrm{i}\Delta$ and $B = -\mathrm{i}V$ applied to terms which are composed of the evolution operator $\mathrm{e}^{t(A+B)}$ associated with the problem and of the evolution operators $\mathrm{e}^{tA}, \mathrm{e}^{tB}$ arising in the splitting scheme; for notational simplicity, we omit an additional scaling factor in the definition of A. Due to the linearity of the problem, the regularity of the initial state and the smoothness of the potential determine the regularity properties of the exact solution, and the regularity of the initial state is inherited by the involved evolution operators, see also [10, Lemma 12]. In order to extend the a priori and a posteriori local error estimates given in [10, Theorems 1,2] to higher-order schemes, a main ingredient are bounds for higher-order iterated commutators. Auxiliary results given for instance in [8, 13], see also references therein, ensure that the k th-order commutator $\mathrm{ad}_A^k(B)\,\psi$ is bounded in terms of $\|V\|_{\mathcal{C}^{2k}}$ and $\|\psi\|_{H^k}$. To keep this presentation self-contained, we briefly explicate these smoothness requirements. A straightforward calculation yields

$$\big[\Delta, V\big]\,\psi = \Delta(V\,\psi) - V\,\Delta\psi = 2\,\nabla V \cdot \nabla\psi + \Delta V\,\psi\,;$$

we point out that the terms comprising second spatial derivatives of ψ cancel. This in particular implies that the first commutator $[A, B] = [\Delta, V]$ is well-defined and bounded for arguments $\psi \in H^1$, provided that the potential satisfies $V \in \mathcal{C}^2$. Similarly, it is seen that the second iterated commutators

$$\big[\Delta, [\Delta, V]\big]\,\psi = 2\,\big[\Delta, \nabla V \cdot \nabla\big]\,\psi + \big[\Delta, \Delta V\big]\,\psi$$

is well-defined for $\psi \in H^2$ and $V \in \mathcal{C}^4$, whereas

$$\big[V, [\Delta, V]\big]\,\psi = 2\,\big[V, \nabla V \cdot \nabla\big]\,\psi + \big[V, \Delta V\big]\,\psi = -\,2\,(\nabla V \cdot \nabla V)\,\psi$$

only requires $\psi \in L^2$ and $V \in \mathcal{C}^1$. By induction, it follows that the dominant error term involving the iterated commutator $\mathrm{ad}_A^k(B)$ is well-defined provided $\psi \in H^k$ and $V \in \mathcal{C}^{2k}$, see also [8, Section 2.2] for detailed arguments.

Altogether, we obtain the following result on the asymptotical correctness of the defect-based a posteriori local error estimators (7), and along the way we recover the known a priori error bounds for higher-order splitting methods.

Theorem 1. *The following local error estimates are valid for an exponential operator splitting method* (3) *of (nonstiff) order $p \geq 1$ applied to the linear Schrödinger equation* (1)*.*

(i) *A priori: If $V \in \mathcal{C}^{2p}$ and $\|\psi_0\|_{H^p} \leq M_p$, then there holds*

$$\|\mathcal{L}(t)\, \psi_0\|_{L^2} \leq C\, t^{p+1}$$

with a constant $C > 0$ depending in particular on M_p.

(ii) *A posteriori: If $V \in \mathcal{C}^{2p+2}$ and $\|\psi_0\|_{H^{p+1}} \leq M_{p+1}$, then the application of the a posteriori local error estimator $\mathcal{P}(t)\, \psi_0$ is well-defined in L^2 and there holds*

$$\|(\mathcal{P} - \mathcal{L})(t)\, \psi_0\|_{L^2} \leq C\, t^{p+2}$$

with a constant $C > 0$ depending in particular on M_{p+1}.

8. Numerical examples

In this section, we illustrate the error behavior of higher-order defect-based local error estimators (7) when applied to test problems of Schrödinger and parabolic type. The numerical results in particular confirm the asymptotical correctness of the constructed a posteriori local error estimators (7).

Splitting methods. For the sake of completeness we specify the coefficients of the employed higher-order splitting methods in Table 1 (real coefficients) and Table 2 (complex coefficients), see also [1, 2]. In addition, we apply the first-order Lie–Trotter splitting method, where $a_1 = b_1 = 1$, and the second-order Strang splitting method, where $a_1 = a_2 = \frac{1}{2}, b_1 = 1, b_2 = 0$.

Test problem of Schrödinger type. As a first illustration, we consider the time-dependent linear Schrödinger equation (1) in one space dimension, subject to the periodic potential $V(x) = \sin^2(\frac{\pi}{4}x)$ and the initial condition $\psi_0(x) = \mathrm{e}^{-x^2+\mathrm{i}\,\sigma_0(x)}$ with $\sigma_0(x) = -\ln\left(\mathrm{e}^{x}+\mathrm{e}^{-x}\right)$. For the space discretization, we apply fast Fourier transform techniques. We truncate the unbounded spatial domain to the interval $[-8, 8]$ and subdivide into $M = 256$ equidistant grid points. Due to the fact that the exact solution remains localized on the considered time interval $[0, 1]$ and as the number of basis functions is chosen sufficiently high, the effect from the artificial periodic boundary conditions and the influence of the spatial error is negligible. For the time integration,

Table 1: Splitting methods with real coefficients of orders $p = 3$ (top), $p = 4$ (middle), and $p = 6$ (bottom).

j	a_j	j	b_j
1	1	1	$-1/24$
2	$-2/3$	2	$3/4$
3	$2/3$	3	$7/24$

j	a_j	j	b_j
1	0	1	0.675603595979829
2	1.351207191959658	2	−0.175603595979829
3	−1.702414383919316	3	−0.175603595979829
4	1.351207191959658	4	0.675603595979829

j	a_j	j	b_j
1	0	1	0.392256805238779
2	0.784513610477557	2	0.510043411918458
3	0.235573213359358	3	−0.471053385409756
4	−1.177679984178871	4	0.068753168252520
5	1.315186320683911	5	0.068753168252520
6	−1.177679984178871	6	−0.471053385409756
7	0.235573213359358	7	0.510043411918458
8	0.784513610477557	8	0.392256805238779

we apply different splitting methods of orders $p = 1, 2, 3, 4, 6$ with real coefficients, namely, the first-order Lie–Trotter splitting, the second-order Strang splitting, a three-stage third-order splitting, a four-stage fourth-order splitting proposed by Yoshida, and an eight-stage sixth-order splitting proposed by Yoshida, see Table 1. A numerical reference solution is computed by a fourth-order splitting scheme proposed in [11], applied with constant time

Table 2: Splitting methods with complex coefficients of orders $p = 3$ (top) and $p = 4$ (bottom).

j	a_j	j	b_j
1	$0.162198202010086 + 0.067293136245403\,\mathrm{i}$	1	$0.415770154056105 + 0.212948225747424\,\mathrm{i}$
2	$0.405225180733310 + 0.198864212461903\,\mathrm{i}$	2	$0.385509228205624 - 0.110555709201699\,\mathrm{i}$
3	$0.432576617256604 - 0.266157348707306\,\mathrm{i}$	3	$0.198720617738271 - 0.102392516545726\,\mathrm{i}$

j	a_j	j	b_j
1	0	1	$0.162198202010086 + 0.067293136245403\,\mathrm{i}$
2	$0.324396404020171 + 0.134586272490807\,\mathrm{i}$	2	$0.337801797989914 - 0.067293136245403\,\mathrm{i}$
3	$0.351207191959658 - 0.269172544981613\,\mathrm{i}$	3	$0.337801797989914 - 0.067293136245403\,\mathrm{i}$
4	$0.324396404020171 + 0.134586272490807\,\mathrm{i}$	4	$0.162198202010086 + 0.067293136245403\,\mathrm{i}$

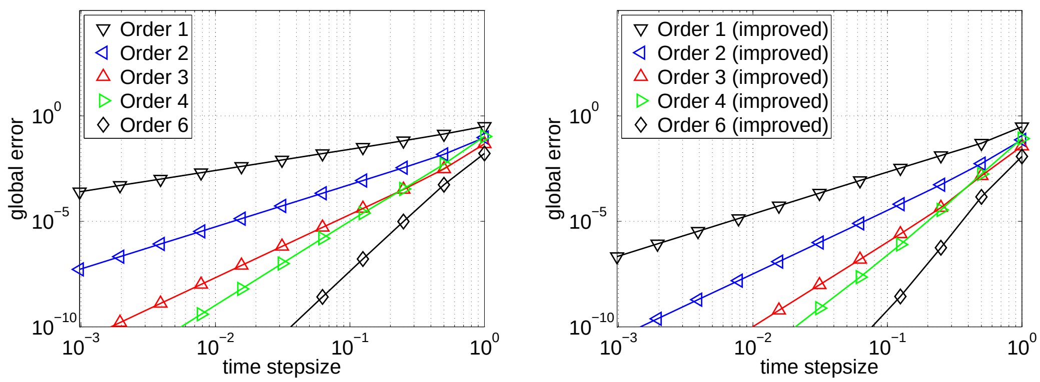

Figure 1: Linear Schrödinger equation: Global errors versus time stepsizes for different splitting methods (left) of orders $p = 1, 2, 3, 4, 6$ and associated improved approximations (right) of orders $p + 1$ resulting from the a posteriori local error estimators (7).

stepsize $\Delta t = 2^{-11}$. The asymptotical correctness of the associated a posteriori local error estimators ensures that a numerical approximation of order $p+1$ is obtained when subtracting the a posteriori local error estimator from the basic solution, since

$$(\mathcal{S} - \mathcal{P}) - \mathcal{E} = \mathcal{L} - \mathcal{P} = \mathscr{O}(t^{p+2})\,.$$

In Figure 1, the global errors of the basic splitting methods and of the associated improved integrators are displayed; the numerical results indeed confirm order $p + 1$ for the improved approximations. Thus, the observed error behavior for the linear test problem of Schrödinger type is in accordance with our theoretical analysis, see Theorem 1.

Test problem of parabolic type. As a further illustration we consider the linear evolution equation (1), but with imaginary unit replaced by one, which leads to a problem of parabolic type. Although this problem class is not strictly covered by our theory, the computations demonstrate that our error analysis also extends to more general classes of problems, provided that the evolution operators associated with the subproblems remain bounded on the involved function spaces. For the time integration we apply the first-order Lie–Trotter splitting, the second-order Strang splitting, a three-stage third-order splitting involving complex coefficients with positive real parts, and a four-stage fourth-order splitting involving complex coefficients with positive

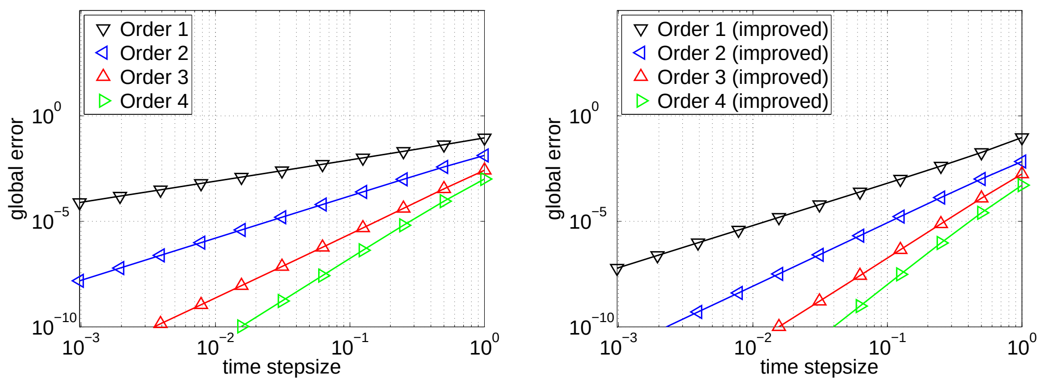

Figure 2: Parabolic equation: Global errors versus time stepsizes for different splitting methods (left) of orders $p = 1, 2, 3, 4$ and associated improved approximations (right) of orders $p + 1$ resulting from the a posteriori local error estimators (7).

real parts, see Table 2 and [12] as well as references given therein. The global errors of the basic splitting methods and of the associated improved integrators, displayed in Figure 2, illustrate the asymptotical correctness of the a posteriori local error estimators.

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