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Efficient Splitting Methods Based on Modified Potentials: Numerical Integration of Linear Parabolic Problems and Imaginary Time Propagation of the Schrödinger Equation

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Abstract. We present a new family of fourth-order splitting methods with positive coefficients especially tailored for the time integration of linear parabolic problems and, in particular, for the time dependent Schrödinger equation, both in real and imaginary time. They are based on the use of a double commutator and a modified processor, and are more efficient than other widely used schemes found in the literature. Moreover, for certain potentials, they achieve order six. Several examples in one, two and three dimensions clearly illustrate the computational advantages of the new schemes.

AMS subject classifications: 65L05, 65M12, 65J10

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1 Introduction

The eigenvalue problem for the stationary Schrödinger equation constitutes an important part in the understanding of basic atomic and molecular phenomena. It is defined by

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($\hbar = m = 1$)

$$\begin{aligned} &\hat{H}\phi_j(x) = E_j\phi_j(x), \qquad j = 0,1,2,\cdots, \\ &\hat{H} = \hat{T} + \hat{V}(x) = -\frac{1}{2}\Delta + \hat{V}(x), \end{aligned} \tag{1.1}$$

where $\hat{V}$ is the potential energy operator and Δ is the Laplacian, an unbounded differential operator. Since the Hamiltonian $\hat{H}$ is Hermitian, then its eigenvalues E_j are real, and the corresponding eigenfunctions ϕ_j can be chosen to form a real orthonormal basis on their domain. By an appropriate election of the origin of the potential we can guarantee that $\hat{V}(x) \geq 0$ in the region of interest, so that $0 \leq E_0 \leq E_1 \leq \cdots$. Given the time-dependent Schrödinger equation

$$i\frac{\partial}{\partial t}\psi(x,t) = \hat{H}\psi(x,t), \quad \psi_0(x) = \psi(x,0), \tag{1.2}$$

if the initial wave function $\psi_0(x)$ is expanded in the orthonormal basis of eigenfunctions ϕ_j,

$$\psi_0(x) = \sum_{j\geq 0} c_j\phi_j(x), \quad c_j = \langle \phi_j(x) \,|\, \psi(x,0)\rangle,$$

where $\langle \cdot | \cdot \rangle$ is the usual L^2-scalar product, then the solution is given by [22]

$$\psi(x,t) = \mathrm{e}^{-it\hat{H}}\psi(x,0) = \sum_{j\geq 0} \mathrm{e}^{-itE_j} c_j\phi_j(x) \tag{1.3}$$

and, in particular, the norm of the solution is preserved for any value of t.

Very often, the so-called imaginary time propagation (ITP) method is the preferred option for solving the eigenvalue problem (1.1) [3, 5, 19] as well as for carrying out path integral simulations in condensed phase quantum systems [16]. By considering the time transformation $t = -i\tau$, Eq. (1.2) is transformed into

$$\frac{\partial}{\partial \tau}\psi(x,\tau) = -\hat{H}\psi(x,\tau), \quad \psi_0(x) = \psi(x,0). \tag{1.4}$$

In this case the solution reads

$$\psi(x,\tau) = \mathrm{e}^{-\tau\hat{H}}\psi(x,0) = \sum_{j\geq 0} \mathrm{e}^{-\tau E_j} c_j\phi_j(x). \tag{1.5}$$

Notice that, in contrast with (1.3), for sufficiently large τ one gets $\psi(x,\tau) \to \mathrm{e}^{-\tau E_0} c_0\phi_0$, since the other exponentials decay more rapidly. In other words, any given wave function at $\tau = 0$ in which $c_0 \neq 0$ converges towards the ground state solution when $\tau \to \infty$. Once an accurate approximation to ϕ_0 is obtained, the associated eigenvalue E_0 can be easily obtained by computing $E_0 = \langle \phi_0(x) | \hat{H} | \phi_0(x) \rangle$. Other functions ϕ_j can also be approximated, e.g., by propagating different wave functions simultaneously in time [2].

To carry out practical computations, it is assumed that $x \in [a_1,b_1]\times\cdots\times[a_k,b_k]$ with each interval $[a_j,b_j]$ sufficiently large so that the wave function and all its derivatives of interest vanish at the boundaries. In this case, we can safely consider periodic boundary conditions and use a pseudo-spectral space discretization. When this is done, one ends up with a finite-dimensional vector $u(\tau)$ approximating the wave function, in the sense that its components $u_j(\tau)\approx\psi(x_j,\tau)$. Then, the problem to be solved corresponds to the linear ODE

$$\begin{cases} u'(\tau) = -\big(T+V\big)\,u(\tau), \quad \tau\in[0,\tau_f], \\ u(0) \text{ given}, \end{cases} \tag{1.6}$$

with solution

$$u(\tau) = \mathrm{e}^{-\tau(T+V)}u(0), \quad t\in[0,\tau_f]. \tag{1.7}$$

Here T and V are the matrices obtained after space discretization approximating the kinetic energy $\hat{T}$ and potential $\hat{V}$, respectively.

A common approach for the time integration of the linear evolution equation (1.6) in the interval $[0,\tau_f]$ is to compute approximations to the exact solution values at certain time grid points by a one-step recurrence:

$$\begin{aligned} &0=\tau_0<\tau_1<\cdots<\tau_N=\tau_f, \\ &u_{n+1}=\mathcal{S}(h_n)\,u_n \approx u(\tau_{n+1}) = \mathrm{e}^{-h_n(T+V)}u(\tau_n), \quad n=0,1,2,\cdots, \end{aligned}$$

where $h_n=\tau_{n+1}-\tau_n$. For simplicity, we assume that $\tau_j=j\Delta\tau$. In other words, we use a constant step size $h_n=h\equiv\Delta\tau$ along the integration. Notice that, since V is diagonal, the computation of $\mathrm{e}^{-\tau V}$ is trivial, whereas $\mathrm{e}^{-\tau T}$ can be done efficiently with the Fast Fourier Transform (FFT). It makes sense, then, to consider splitting methods to approximate the exact evolution, the Strang splitting being a prototypical example:

$$\mathcal{S}^{[2]}(h)\,u_0 \equiv \mathrm{e}^{-\frac{h}{2}V}\mathrm{e}^{-hT}\mathrm{e}^{-\frac{h}{2}V}u_0. \tag{1.8}$$

It is verified that $\mathcal{S}^{[2]}(h)=\mathrm{e}^{-h(T+V)}+\mathcal{O}(h^3)$. Since the problem is assumed to be periodic, each operator (or exponential) satisfies the boundary conditions and no order reduction occurs. Moreover, this scheme can be used for integrating the Schrödinger equation both in real time and in imaginary time because $\|\mathrm{e}^{-\frac{h}{2}V}\|\leq 1$ and $\|\mathrm{e}^{-hT}\|\leq 1$.

Higher order approximations (say, of order $p\geq 3$) can be achieved by a more general composition of the form

$$\mathcal{S}^{[p]}(h)\,u_0 = \mathrm{e}^{-hb_{s+1}V}\mathrm{e}^{-ha_{s+1}T}\cdots\mathrm{e}^{-hb_1V}\mathrm{e}^{-ha_1T}u_0, \tag{1.9}$$

where the coefficients a_j, b_j are chosen so that $\mathcal{S}^{[p]}(h)=\mathrm{e}^{-h(T+V)}+\mathcal{O}(h^{p+1})$. Splitting methods of this form have also been recently applied in different areas, as shown e.g. in the contributions [1,11,14].

Notice that $\|\mathrm{e}^{-a_ihT}\|$ is bounded only if $a_i>0$, whereas $\|\mathrm{e}^{-b_ihV}\|$ is bounded in the domain of interest even if $b_i<0$, although in regions where the potential takes large

values it can take exponentially large values and significant roundoff errors may occur. It is therefore desirable to have all coefficients $b_i > 0$, otherwise one has to incorporate an upper cut off of the potential to reduce such errors.

In this respect, a well known result establishes that there are no splitting methods of the form (1.9) of order greater than two (i.e., $p > 2$) with all their coefficients being real and positive. Thus, if one is interested in applying splitting methods of order 3 or higher with real coefficients, then at least one a_j and one b_k have to be negative [15, 28, 30] (a simple proof can be found in [6]). Whereas this is usually not a problem if the splitting method is applied to the Schrödinger equation in real time (1.2), the presence of negative a_i coefficients makes the ITP algorithm badly conditioned. This feature can be traced back to the fact that (1.4) is a parabolic equation involving unbounded negative definite operators, and therefore the solution evolves in a semigroup. On the other hand, when the methods are applied to the usual Schrödinger equation in real time, the presence of only positive coefficients leads to methods with very good stability properties.

One possible way to overcome this order barrier consists in including the gradient of the potential, $\nabla V(x)$, in the composition. In fact, several fourth-order forward methods involving the gradient of the potential exist in the literature and some of them have been successfully used in this context [12, 17, 23]. Not all of them, however, can be used for parabolic equations or have good stability properties and/or efficiency.

In this work we propose new splitting methods involving $\nabla V(x)$ in their formulation and only positive coefficients. They are primarily aimed to be applied in the ITP algorithm, but are also valid for the numerical integration in time of the initial value problem originated when discretizing more general linear parabolic problems in space. The paper is structured as follows. In Section 2 we review the most efficient 4th-order splitting methods with modified potentials and positive coefficients we have found in the literature. In particular, the authors of [24] present a large collection of schemes involving up to five stages. Whereas their main objective was just getting efficient schemes, irrespective of the sign of the coefficients, it turns out that most of the integrators do contain only positive coefficients. Here we collect and test the most efficient among them.

Typically, when one increases the number of exponentials in a splitting method it is possible to reduce the leading error terms in such as way that this reduction makes up for the extra cost involved. In this case, however, due to the existing order barrier for methods with positive coefficients, the overall improvement in accuracy hardly compensates the extra cost when additional stages are included. To deal with this problem, we present in Section 3 a novel procedure leading to more efficient schemes. It is based on a conveniently modified processing of a basic method involving a reduced number of stages. The new splitting methods are tested in Section 4 on several numerical examples in comparison with the most efficient integrators from the literature. Finally, Section 5 contains some concluding remarks.

2 A review of operator splitting methods

2.1 Standard splitting methods

The general splitting method (1.9)

$$\mathcal{S}^{[p]}(h)=\prod_{j=1}^{s+1}\mathrm{e}^{-hb_jV}\,\mathrm{e}^{-ha_jT}, \tag{2.1}$$

will be written here in a more abbreviated form just by enumerating the sequence of its coefficients,

$$[b_{s+1}a_{s+1}\cdots b_2a_2b_1a_1]. \tag{2.2}$$

Moreover, we will only consider time-symmetric methods, corresponding to left-right palindromic compositions. In other words, we have in (2.2) either

$$b_{s+1}=0 \quad \text{and} \quad a_{s+2-j}=a_j,\ b_{s+1-j}=b_j \tag{2.3}$$

or

$$a_1=0 \quad \text{and} \quad b_{s+2-j}=b_j,\ a_{s+2-j}=a_{j+1}, \tag{2.4}$$

$j=1,2,\cdots,s$. In the first case the compact form of the method reads

$$[a_1b_1a_2b_2\cdots b_2a_2b_1a_1], \tag{2.5}$$

whereas a method verifying (2.4) will be denoted for simplicity by[†]

$$[b_1a_1b_2a_2\cdots a_2b_2a_1b_1]. \tag{2.6}$$

Thus, in particular, the Strang splitting (1.8) reads $[\frac{1}{2}1\frac{1}{2}]$.

In both cases, s is referred to as the *number of stages* of the integrator[‡]. Notice that, due to the different character of the operators T and V, the role of e^{-hT} and e^{-hV} is not interchangeable in (2.1), and thus these two different orderings have to be analyzed. We will refer to (2.5) and (2.6) as methods of type ABA and BAB, respectively.

2.2 Modified splitting methods

An essential observation with respect to the operators $\hat{T}$ and $\hat{V}$ in (1.1) is that they verify the relation

$$[\hat{V},[\hat{T},\hat{V}]]\psi=|\nabla\hat{V}|^2\psi \tag{2.7}$$

[†]Strictly speaking, the sequence is $[b_1a_2b_2a_3\cdots a_3b_2a_2b_10]$, but we remove the zero coefficient and shift the index of the a_i coefficients for clarity.

[‡]The last map can be concatenated with the first one in the following step and so it is not counted for the cost of the method.

for any ψ, where $[\hat{T},\hat{V}]=\hat{T}\hat{V}-\hat{V}\hat{T}$. Since the double commutator $[\hat{V},[\hat{T},\hat{V}]]$ is only a function of $\hat{V}$, it is usually referred to as a *modified potential*. Furthermore, $[\hat{V},[\hat{V},[\hat{T},\hat{V}]]]=0$. Similar relations hold for the matrices T and V resulting from the (sufficiently accurate) space discretization of $\hat{T}$ and $\hat{V}$, respectively, with $[V,[T,V]]$ diagonal if the derivatives of the potential are computed first and then evaluated on the corresponding space grid. In consequence, we can replace the terms $\exp(-hb_jV)$ in the scheme (2.1) by the more general operator $\exp(-hb_jV-h^3c_j[V,[T,V]])$, involving two parameters. More formally, we can take compositions of the form

$$\mathcal{S}_{\rm mod}(h)u_0=\prod_{j=1}^{s+1}\mathrm{e}^{-hb_jV-h^3c_j[V,[T,V]]}\mathrm{e}^{-ha_jT}u_0. \tag{2.8}$$

Such integrators are called *modified operator splitting methods*, and will be denoted as

$$\begin{aligned}&\text{ABA:}\quad [a_{s+1}(b_s,c_s)\cdots(b_2,c_2)a_2(b_1,c_1)a_1],\\ &\text{BAB:}\quad [(b_{s+1},c_{s+1})a_s\cdots(b_2,c_2)a_1(b_1,c_1)].\end{aligned}$$

Methods of this class have been used in different situations, starting from the pioneering work of Ruth [26]. Early references also include [25, 31–33, 35]. It is indeed possible to get fourth-order methods with positive coefficients [12, 17, 24], and in fact one of the most popular schemes corresponds to the composition [12, 17]

$$\mathcal{S}^{[4]}(h)\equiv\mathrm{e}^{-\frac{h}{6}V}\mathrm{e}^{-\frac{h}{2}T}\mathrm{e}^{-\frac{2h}{3}V-\frac{h^3}{72}[V,[T,V]]}\mathrm{e}^{-\frac{h}{2}T}\mathrm{e}^{-\frac{h}{6}V}. \tag{2.9}$$

Although methods up to order eight have been designed [24], they involve some negative coefficients a_i when the order is higher than 4. It has been argued that the maximal order one can achieve with all a_j real and positive is indeed 4 [4, 13]. On the other hand, there are some 6th-order methods with all b_i coefficients positive. In any case, these methods cannot be applied in the context of parabolic equations involving the Laplacian.

When analyzing operator splitting methods, and in particular their truncation errors and efficiencies, it is a common practice to express compositions (2.1) and (2.8) as $\mathcal{S}(h)=\exp(-F(h))$ by means of the Baker–Campbell–Hausdorff formula [34]. Specifically,

$$F(h)=h(f_{1,1}T+f_{1,2}V)+h^3\left(f_{3,1}E_{3,1}+f_{3,2}E_{3,2}\right)+h^5\sum_{j=1}^{4}f_{5,j}E_{5,j}+\mathcal{O}(h^7) \tag{2.10}$$

when the compositions are palindromic. Here the $f_{k,j}$ are polynomial functions depending on the coefficients of the scheme and $E_{k,j}$ denote the elements of a basis of the homogeneous subspace $\mathcal{L}^k(T,V)$ of grade k of the Lie algebra $\mathcal{L}(T,V)$ generated by the operators T and V with the commutator as the Lie bracket. The specific basis for $k\leq 5$ used in this work is given in Table 1.

A method is of order four if $f_{1,1}=f_{1,2}=1$ (for consistency) and $f_{3,1}=f_{3,2}=0$, whereas the quantity $\left(\sum_{j=1}^{4}f_{5,j}^2\right)^{1/2}$ is usually taken as a measure of its error. This is multiplied by

Table 1: Elements $E_{k,j}$ of the basis of the homogeneous subspace $\mathcal{L}^k(T,V)$ for $k\leq 5$.

$k=1$	$E_{1,1}=T \quad E_{1,2}=V$
$k=2$	$E_{2,1}=[T,V]$
$k=3$	$E_{3,1}=[T,[T,V]] \quad E_{3,2}=[V,[T,V]]$
$k=4$	$E_{4,1}=[T,E_{3,1}] \quad E_{4,2}=[T,E_{3,2}]$
$k=5$	$E_{5,1}=[T,E_{4,1}] \quad E_{5,2}=[V,E_{4,1}] \quad E_{5,3}=-[T,E_{4,2}]$ $E_{5,4}=[V,E_{4,2}]$

the number of stages s to take also into account the computational cost, so that one can compare the efficiency of methods with different stages by taking

$$\mathcal{E}_f = s^4 \cdot \left(\sum_{j=1}^{4} f_{5,j}^2 \right)^{1/2} \tag{2.11}$$

as an estimate of the effective error of a 4th-order scheme.

As stated before, there are in the literature a number of 4th-order integrators with positive coefficients within the family (2.8). Among them, we have identified those with the smallest truncation error terms. They are collected according to the number stages they involve, whereas the corresponding values of $|f_{5,j}|$ and their effective errors $\mathcal{E}_f$ are gathered in Table 2.

$s=1$. The simplest scheme is of course obtained by incorporating the double commutator $[V,[T,V]]$ into the Strang splitting method (1.8), namely

$$\mathcal{S}_{\mathrm{R}_1}(h) = \mathrm{e}^{-\frac{h}{2}V-\frac{h^3}{48}E_{3,2}}\mathrm{e}^{-hT}\mathrm{e}^{-\frac{h}{2}V-\frac{h^3}{48}E_{3,2}} \tag{2.12}$$

for a BAB method and

$$\mathcal{S}_{\mathrm{R}_2}(h) = \mathrm{e}^{-\frac{h}{2}T}\mathrm{e}^{-hV-\frac{h^3}{24}E_{3,2}}\mathrm{e}^{-\frac{h}{2}T} \tag{2.13}$$

for an ABA scheme. Methods (2.12) and (2.13) do not contain enough parameters to achieve order four, but in both cases the parameter multiplying the double commutator is chosen so as to satisfy the condition $f_{3,1}=f_{3,2}$. This, as we will see, leads to schemes of effective order four, in the sense that, by applying an appropriate near-identity transformation, one gets a method of order 4 [25,33,35].

$s=2$. Scheme (2.9) belongs indeed to this class. In our compact notation, it reads

$$[b_1 a_2(b_2,c_2) a_2 b_1] \equiv \left[\frac{1}{6}\frac{1}{2}\left(\frac{2}{3},\frac{1}{72}\right)\frac{1}{2}\frac{1}{6}\right]. \tag{2.14}$$

A similar ABA composition exists,

$$[a_1(b_1,c_1)a_2(b_1,c_1)a_1] \tag{2.15}$$

Table 2: Main truncation error terms and effective error for standard splitting operator methods of order 4 with positive coefficients and s stages.

Type	s	$\lvert f_{5,1}\rvert$	$\lvert f_{5,2}\rvert$	$\lvert f_{5,3}\rvert$	$\lvert f_{5,4}\rvert$	$\mathcal{E}_f$
ABA	2	$2.31\cdot10^{-4}$	$1.57\cdot10^{-4}$	$4.58\cdot10^{-4}$	$4.71\cdot10^{-4}$	$1.14\cdot10^{-2}$
	3	$8.82\cdot10^{-6}$	$7.05\cdot10^{-6}$	$3.59\cdot10^{-6}$	$3.17\cdot10^{-6}$	$9.94\cdot10^{-4}$
	4	$2.07\cdot10^{-6}$	$1.96\cdot10^{-6}$	$8.87\cdot10^{-7}$	$8.61\cdot10^{-7}$	$7.97\cdot10^{-4}$
BAB	2	$3.47\cdot10^{-4}$	$4.63\cdot10^{-4}$	$3.47\cdot10^{-4}$	$2.31\cdot10^{-4}$	$1.14\cdot10^{-2}$
	3	$1.09\cdot10^{-5}$	$1.13\cdot10^{-5}$	$4.29\cdot10^{-6}$	$3.71\cdot10^{-6}$	$1.35\cdot10^{-3}$
	4	$2.46\cdot10^{-6}$	$2.67\cdot10^{-6}$	$4.08\cdot10^{-7}$	$3.84\cdot10^{-7}$	$9.42\cdot10^{-4}$

with

$$a_1=\frac{1}{2}-\frac{1}{2\sqrt{3}},\quad b_1=\frac{1}{2},\quad c_1=-\frac{1}{24}+\frac{\sqrt{3}}{48},\quad a_2=\frac{1}{\sqrt{3}},$$

but it exhibits a slightly worse performance in practice, and $c_1<0$.

More efficient schemes can be achieved by introducing additional stages. Thus, in reference [24] different compositions with 3, 4 and 5 stages of types (2.3) and (2.4) are presented, many of them having all coefficients $a_j>0$. Among them, we collect those recommended in [24] as the most efficient.

$s=3$. Specifically, the coefficients of the ABA and BAB methods are, respectively

$$\begin{array}{ll}
[a_1(b_1,c_1)a_2(b_2,c_2)a_2(b_1,c_1)a_1] & [(b_1,c_1)a_1(b_2,c_2)a_2(b_2,c_2)a_1(b_1,c_1)] \\
========== & ========== \\
a_1=0.1159953608486416 & b_1=0.08002565306418866 \\
b_1=0.2825633404177051 & c_1=0.0002725753410753895 \\
c_1=0.00122608898953636 1 & a_1=0.2728983001988755 \\
a_2=\tfrac{1}{2}-a_1 & b_2=\tfrac{1}{2}-b_1 \\
b_2=1-2b_1 & c_2=0.002960781208329478 \\
c_2=0.003035236056708454 & a_2=1-2a_1
\end{array} \tag{2.16}$$

$s=4$. The coefficients of the most efficient ABA and BAB methods are

$$\begin{array}{ll}
[a_1(b_1,c_1)a_2(b_2,c_2)a_3(b_2,c_2)a_2(b_1,c_1)a_1] & [(b_1,c_1)a_1b_2a_2(b_3,c_3)a_2b_2a_1(b_1,c_1)] \\
========== & ========== \\
a_1=0.09325912861071900 & b_1=0.05851872613455621 \\
b_1=0.2247800288685984 & c_1=0.4339598806816256\cdot10^{-3} \\
c_1=0.0005562281089130940 & a_1=0.1921125277429464 \\
a_2=0.2791634819768266 & b_2=0.2852162240687091 \\
b_2=\tfrac{1}{2}-b_1 & a_2=\tfrac{1}{2}-a_1 \\
c_2=0.0008405927247441154 & b_3=1-2(b_1+b_2) \\
a_3=1-2(a_1+a_2) & c_3=0.2427475259663050\cdot10^{-2}
\end{array} \tag{2.17}$$

Several methods with 5 stages have also been reported but either they have larger effective error or at least one of the a_j coefficients is negative.

For special classes of problems, such as near-integrable systems, there are particularly efficient schemes within this class when the perturbation is small. Some examples can be found in [5, 21, 29], but they will not be considered in this work.

3 Modified processed splitting methods

3.1 Methods with processing

Given a method $\mathcal{S}(h)$, one may consider a near-to-identity transformation, $\mathcal{P}(h)=\mathcal{I}+\mathcal{O}(h^q)$ with $q\geq 1$, so that the composition

$$\widehat{\mathcal{S}}(h)=\mathcal{P}(h)^{-1}\cdot\mathcal{S}(h)\cdot\mathcal{P}(h) \tag{3.1}$$

is more accurate than $\mathcal{S}(h)$, for instance by increasing its order, whereas still possessing the same stability as $\mathcal{S}(h)$. This is the case, in particular, of method (2.13): by taking $\mathcal{P}(h)=\mathrm{e}^{\frac{1}{24}h^2[T,V]}$ one ends up with a 4th-order integrator. In this setting, $\mathcal{S}(h)$ is called the *kernel* of the processed method $\widehat{\mathcal{S}}(h)$, and $\mathcal{P}(h)$ is the *processor* or *corrector* [8–10]. Notice that N consecutive steps of the processed integrator correspond to the transformation

$$\begin{aligned}\widehat{\mathcal{S}}(h)^N&=\overbrace{\widehat{\mathcal{S}}(h)\cdots\widehat{\mathcal{S}}(h)}^{N\text{times}}=\overbrace{(\mathcal{P}(h)^{-1}\cdot\mathcal{S}(h)\cdot\mathcal{P}(h)\cdots\mathcal{P}(h)^{-1}\cdot\mathcal{S}(h)\cdot\mathcal{P}(h)}^{N\text{times}})\\&=\mathcal{P}(h)^{-1}\cdot\mathcal{S}(h)^N\cdot\mathcal{P}(h).\end{aligned} \tag{3.2}$$

Thus, to perform N steps of the processed method, one successively (i) applies once the map $\mathcal{P}(h)$ (preprocessing), (ii) takes N steps of the kernel $\mathcal{S}(h)$ and finally (iii) applies the map $\mathcal{P}(h)^{-1}$ (postprocessing). Since $\mathcal{P}(h)$ and its inverse are applied only once, the computational complexity of $\widehat{\mathcal{S}}(h)$ corresponds essentially to that of $\mathcal{S}(h)$ if N is sufficiently large. The kernel $\mathcal{S}(h)$ is said to be of effective order p if a processor can be constructed leading to a method of genuine order p.

When the kernel is time-symmetric, the processor leading to a minimum main order truncation error must be such that $\mathcal{P}(-h)=\mathcal{P}(h)$. With this requirement, the whole method is also time-symmetric, i.e. $\widehat{\mathcal{S}}(h)^{-1}=\widehat{\mathcal{S}}(-h)$ [7].

Of course, this "ideal" processor (e.g. $\mathcal{P}(h)=\mathrm{e}^{\frac{1}{24}h^2[T,V]}$ in the previous case) is, in general, computationally involved and it will only be approximated in practical applications. This is done once again with a product of the form [7]

$$\Pi(h)=\prod_{j=1}^{\ell}\mathrm{e}^{-h\beta_jV}\,\mathrm{e}^{-h\alpha_jT} \tag{3.3}$$

for certain coefficients α_j, β_j, $j=1,\cdots,\ell$, chosen so that

$$\Pi(h)=\mathcal{P}(h)+\mathcal{O}(h^r)$$

with $r \geq p$, the order of the method itself. In this way, the overall method

$$\Pi(h)^{-1} \cdot \mathcal{S}(h) \cdot \Pi(h) \tag{3.4}$$

has the same structure as the kernel, since

$$\Pi(h)^{-1} = \mathrm{e}^{h\alpha_1 T} \mathrm{e}^{h\beta_1 V} \cdots \mathrm{e}^{h\alpha_\ell T} \mathrm{e}^{h\beta_\ell V}.$$

Notice, however, that the resulting scheme (3.4) is no longer time-symmetric (since the composition is not palindromic. Notice also that $\Pi(-h) \neq \Pi(h)$ while the ideal one satisfies that $\mathcal{P}(-h) = \mathcal{P}(h)$) and in addition involves negative coefficients: they must verify

$$\alpha_1 + \cdots + \alpha_\ell = 0, \quad \beta_1 + \cdots + \beta_\ell = 0$$

for consistency (as well as the fact that the coefficients $\pm\alpha_i, \pm\beta_i$ appear either in the pre-processor or in the post-processor).

The first drawback can be overcome by using in (3.4) the *adjoint* of $\Pi(h)$ instead of its inverse. Let us recall that the adjoint $\Pi(h)^*$ is defined as the map such that $\Pi(-h)^* = \Pi(h)^{-1}$ [27]. It is then clear that

$$\Pi(h)^* = \mathrm{e}^{-h\alpha_1 T} \mathrm{e}^{-h\beta_1 V} \cdots \mathrm{e}^{-h\alpha_\ell T} \mathrm{e}^{-h\beta_\ell V},$$

so that

$$\Pi(h)^* = \Pi(-h)^{-1} = \mathcal{P}(-h)^{-1} + \mathcal{O}(h^r) = \mathcal{P}(h)^{-1} + \mathcal{O}(h^r)$$

if the processor satisfies $\mathcal{P}(-h) = \mathcal{P}(h)$. In this way we propose to apply, instead of (3.4), the scheme

$$\widetilde{\mathcal{S}}_N(h) = \Pi(h)^* \cdot \mathcal{S}(h)^N \cdot \Pi(h). \tag{3.5}$$

Since

$$(\widetilde{\mathcal{S}}_N(h))^{-1} = \Pi(h)^{-1} \cdot (\mathcal{S}(h)^N)^{-1} \cdot (\Pi(h)^*)^{-1}$$

and

$$\widetilde{\mathcal{S}}_N(-h) = \Pi(-h)^* \cdot \mathcal{S}(-h)^N \cdot \Pi(-h) = \Pi(h)^{-1} \cdot \mathcal{S}(-h)^N \cdot (\Pi(h)^*)^{-1},$$

then the modified processed integrator (3.5) is also time-symmetric if $\mathcal{S}(h)$ is time-symmetric.

On the other hand, the second difficulty can be dealt with the idea of *starter*. It proceeds essentially as follows. Suppose that our kernel $\mathcal{S}(h)$ is time-symmetric and one is interested in applying $N \geq 2$ steps of the processed scheme. If we set

$$\widehat{\Pi}(h) \equiv \mathcal{S}(h) \cdot \Pi(h),$$

then Eq. (3.5) can be rewritten as

$$\widetilde{\mathcal{S}}_N(h) = \widehat{\Pi}(h)^* \cdot \mathcal{S}(h)^{N-2} \cdot \widehat{\Pi}(h),$$

and finally the map $\widehat{\Pi}(h)$ is approximated as

$$\widehat{\Pi}(h) \simeq \mathrm{e}^{-h\beta_n V}\mathrm{e}^{-h\alpha_n T}\cdots\mathrm{e}^{-h\beta_1 V}\mathrm{e}^{-h\alpha_1 T}, \tag{3.6}$$

for another set of coefficients α_i, β_i. Now consistency requires instead

$$\alpha_1+\cdots+\alpha_n=1, \quad \beta_1+\cdots+\beta_n=1,$$

and these relations may in principle be satisfied with $\alpha_i,\beta_i \geq 0$, $i=1,\cdots,n$, for specific kernels.

It is also clear that we can replace $\exp(-h\beta_j V)$ in the previous procedure by the more general operator $\exp(-h\beta_j - h^3 c_j E_{3,2})$ when necessary.

Since $\widehat{\Pi}(h)$ can also be seen as a one step method, then the order barrier also applies to this family of methods and only methods up to order four with $a_i > 0$ can be obtained.

3.2 Order conditions

To construct specific methods within this class we have to find and solve the corresponding order conditions. This can be done as in standard compositions, i.e., by expressing both the kernel and the processor as the exponential of just one operator. Thus, for the kernel, one has $\mathcal{S}(h)=\exp(-K(h))$, with

$$K(h)=h(T+V)+h^3\big(k_{3,1}E_{3,1}+k_{3,2}E_{3,2}\big)+h^5\sum_{j=1}^{4}k_{5,j}E_{5,j}+\mathcal{O}(h^7) \tag{3.7}$$

and the order conditions to achieve a method of effective order 6 are [8]

$$\begin{aligned} &N_{3,1}=k_{3,1}-k_{3,2}=0,\\ &N_{5,1}=k_{5,2}-k_{5,1}-\frac{1}{2}k_{3,1}^2=0,\\ &N_{5,2}=k_{53}+k_{5,4}-k_{3,1}k_{3,2}=0. \end{aligned} \tag{3.8}$$

With respect to the processor $\Pi(h)$, it can be expressed as $\Pi(h)=\exp(-P(h))$, with

$$P(h)=h^2 p_{2,1}E_{2,1}+h^4\big(p_{4,1}E_{4,1}+p_{4,2}E_{4,2}\big)+\mathcal{O}(h^6).$$

Notice that, for the ideal processor, $p_{2k+1,j}=0$ for $k\geq 0$. In consequence, the processed method reads

$$\Pi(h)^{-1}\cdot\mathcal{S}(h)\cdot\Pi(h)=\exp(-F(h)),$$

where now

$$F(h)=h(T+V)+h^3\big(f_{3,1}E_{3,1}+f_{3,2}E_{3,2}\big)+h^5\sum_{j=1}^{4}f_{5,j}E_{5,j}+\mathcal{O}(h^7) \tag{3.9}$$

and

$$f_{3,1}=k_{3,1}+p_{2,1},\quad f_{3,2}=k_{3,2}+p_{2,1},\quad f_{5,1}=k_{5,1}+p_{4,1},$$
$$f_{5,2}=k_{5,2}+k_{3,1}p_{2,1}+\frac{1}{2}p_{2,1}^2+p_{4,1},\quad f_{5,3}=k_{5,3}+k_{3,1}p_{2,1}+\frac{1}{2}p_{2,1}^2-p_{4,2},$$
$$f_{5,4}=k_{5,4}+k_{3,2}p_{2,1}+\frac{1}{2}p_{2,1}^2p_{4,2}.$$

Accordingly, if $N_{3,1}=0$ and $p_{2,1}=-k_{3,1}$ (in addition to the conditions $p_{1,j}=p_{3,j}=0$), then the processed method is of order four. This is precisely what happens with the methods with one stage (2.12) and (2.13).

Unfortunately, conditions $N_{5,1}=0$ and $N_{5,2}=0$ cannot be simultaneously verified by kernels with positive coefficients: one can still find coefficients c_j such that $N_{5,2}=0$ with positive coefficients, whereas the processor can be used to vanish $f_{5,3}$ and $f_{5,4}$ simultaneously. However, the constraint $a_j>0$ in the kernel implies that $N_{5,1}\neq 0$ and then

$$f_{5,1}=0,\quad f_{5,2}=0$$

cannot be simultaneously satisfied with the processor.

In view of the situation, our proposal here is to choose coefficients for the kernel verifying $N_{3,1}=N_{5,2}=0$ and take $p_{4,1}$ such that one of the following requirements is satisfied:

(a) $f_{5,1}=-N_{5,1}$, $f_{5,2}=0$;

(b) $f_{5,1}=0$, $f_{5,2}=N_{5,1}$;

(c) $f_{5,1}=\frac{1}{2}N_{5,1}$, $f_{5,2}=-\frac{1}{2}N_{5,1}$,

with $N_{5,1}$ as small as possible. In this way the effective error achieves the minimum value in all cases. Alternatives (a) and (b) can be relevant depending on the type of potential and wave function one is considering. For instance, without loss of generality, suppose one is dealing with a one-dimensional problem and denote $V^{(k)}\equiv\frac{d^k\hat{V}(x)}{dx^k}$, and $\psi^{(k)}\equiv\frac{\partial^k\psi(x,t)}{\partial x^k}$, $k\geq 1$. Then, a straightforward computation shows that

$$\hat{E}_{5,1}\psi=V^{(8)}\psi+8V^{(7)}\psi^{(1)}+24V^{(6)}\psi^{(2)}+32V^{(5)}\psi^{(3)}+8V^{(4)}\psi^{(4)},$$
$$\hat{E}_{5,2}\psi=\left(-6V^{(5)}V^{(1)}-12V^{(4)}V^{(2)}+8(V^{(3)})^2\right)\psi+24\left(V^{(3)}V^{(2)}-V^{(4)}V^{(1)}\right)\psi^{(1)}$$
$$+24V^{(3)}V^{(1)}\psi^{(2)},$$

where $\hat{E}_{5,j}$ denotes the corresponding element of Table 1 obtained with the operators $\hat{T}$ and $\hat{V}$. Now, if $\hat{V}(x)$ is at most cubic, then $\hat{E}_{5,1}\psi=0$. In fact, the contribution of this term to the error is almost negligible for smooth potentials or wave functions describing the system near the ground state, or evolving near the minimum of the potential. Then, the choice (a) would lead to a 6th-order method with positive coefficients for cubic potentials.

With respect to the starter, one has

$$\widehat{\Pi}(h)=\exp(-K(h))\cdot\exp(-P(h))=\exp\left(-\sum_{k,j}h^k t_{k,j}E_{k,j}\right),$$

and the previous conditions for the processor to achieve order four lead to

$$\begin{aligned}&t_{1,1}=1,\quad t_{1,2}=1,\\&t_{2,1}=k_{3,1},\\&t_{3,1}=\frac{1}{2}k_{3,1},\quad t_{3,2}=\frac{1}{2}k_{3,1},\end{aligned}\tag{3.10}$$

whereas additional restrictions are necessary for $t_{4,j}$ and $t_{5,j}$ according to the options (a), (b) or (c) above. Thus, in case (a) it is required that

$$\begin{aligned}&t_{4,1}=\frac{1}{12}k_{3,1}-\frac{1}{2}k_{3,1}^2+k_{5,2},\quad t_{4,2}=\frac{1}{6}(k_{3,1}+3k_{3,1}^2-6k_{5,3}),\\&t_{5,1}=k_{5,1}+\frac{1}{4}(k_{3,1}^2-2k_{5,2}),\quad t_{5,2}=\frac{1}{6}(-k_{3,1}^2+3k_{5,2}),\\&t_{5,3}=\frac{1}{6}(-k_{3,1}^2+3k_{5,3}),\quad t_{5,4}=\frac{1}{6}(2k_{3,1}^2-3k_{5,3}).\end{aligned}\tag{3.11}$$

We will take then a composition involving at least 11 parameters to solve all the conditions. In summary, the procedure to construct operator splitting methods with starter is the following:

1. Obtain the coefficients a_j, b_j, c_j of time-symmetric kernels (with $a_j>0$, $b_j>0$) by solving $N_{3,1}=N_{5,2}=0$ and minimizing $N_{5,1}$.
2. Determine the values of the relevant terms $k_{i,j}$ for this kernel and from them, the terms $t_{i,j}$ of the starter.
3. Obtain the coefficients α_j, β_j, γ_j of the composition (3.6) defining the starter (again with $\alpha_j>0$, $\beta_j>0$) according with the chosen alternative (a), (b) or (c).

3.3 Specific methods

We have obtained kernels of type (2.3) and (2.4) involving up to $s=4$ stages by applying the previous methodology. In Table 3 we collect the corresponding values of $|N_{5,1}|$ and, to take into account the computational cost, also the value of $s^4\frac{1}{\sqrt{2}}|N_{5,1}|$ in each case. This last quantity is closely related with the overall effective error of the processed method.

We see that, although the value of $|N_{5,1}|$ can be reduced by increasing the number of stages, this reduction is hardly compensated by the higher computational cost required. For this reason, we restrict ourselves to kernels with only one and two stages.

Table 3: Error terms for the kernel of processed splitting operator methods. When $s=1$, $N_{5,2}=-N_{5,1}$.

Type	s	$\lvert N_{5,1}\rvert$	$\frac{s^4}{\sqrt{2}}\lvert N_{5,1}\rvert$
ABA	1	$6.94\cdot10^{-4}$	$4.91\cdot10^{-4}$
	2	$6.06\cdot10^{-4}$	$6.85\cdot10^{-3}$
	3	$8.57\cdot10^{-6}$	$4.90\cdot10^{-4}$
	4	$3.17\cdot10^{-6}$	$5.73\cdot10^{-4}$
BAB	1	$6.94\cdot10^{-4}$	$4.91\cdot10^{-4}$
	2	$4.44\cdot10^{-5}$	$5.02\cdot10^{-4}$
	3	$8.59\cdot10^{-6}$	$4.91\cdot10^{-4}$
	4	$2.71\cdot10^{-6}$	$4.90\cdot10^{-4}$

$s=1$. In the particular case of kernel (2.13), one has $N_{3,1}=0$, but neither $N_{5,1}$ nor $N_{5,2}$ vanish, so that we apply a different strategy to construct a cheaper starter leading to a method of order 4. It turns out that this can be achieved with

$$\widehat{\Pi}(h)=\left[(\beta_3,\gamma_3)\alpha_3(\beta_2,\gamma_2)\alpha_2(\beta_1,\gamma_1)\alpha_1\right]. \tag{3.12}$$

In fact, only 2 modified potentials are required and still one has a 1-parameter family of solutions. Among them we choose

$$\begin{aligned}
&\alpha_1=0.015910816538916105477, && \alpha_2=0.52240758893355298829,\\
&\beta_1=0.16194613148158516891, && \beta_2=0.82805386851841483109,\\
&\gamma_1=0.006147039752336731864 1, && \gamma_2=0.018617166558200649744,\\
&\alpha_3=0.46168159452753090624, && \beta_3=\tfrac{1}{100},\\
&\gamma_3=0.
\end{aligned}$$

The overall scheme reads

$$\begin{aligned}
\widetilde{\mathcal{S}}_N(h)=&\left[\alpha_1(\beta_1,\gamma_1)\alpha_2(\beta_2,\gamma_2)\alpha_3(\beta_3,\gamma_3)\right]\cdot\left[\frac{1}{2}(1,\frac{1}{24})\frac{1}{2}\right]^{N-2}\\
&\cdot\left[(\beta_3,\gamma_3)\alpha_3(\beta_2,\gamma_2)\alpha_2(\beta_1,\gamma_1)\alpha_1\right].
\end{aligned} \tag{3.13}$$

We should remark that the modified processor has not been chosen to optimize the truncation error, since $N_{5,2}\neq0$ anyway. Moreover, the method has to be used with $N\geq2$ and the total number of stages is $N+4$. In consequence, for small values of N, this extra cost can be significant with respect to the cost of the kernel.

$s=2$. The kernels read respectively

$$\text{ABA}:\qquad \mathcal{S}_{\text{ABA}}(h)=\left[a_1(b_1,c_1)a_2(b_1,c_1)a_1\right] \tag{3.14}$$

and

$$\text{BAB}:\qquad \mathcal{S}_{\text{BAB}}(h)=\left[(b_1,c_1)a_2(b_2,c_2)a_2(b_1,c_1)\right]. \tag{3.15}$$

Once the kernel has been chosen, the starter can be determined according with the previous alternatives (a), (b) and (c), so that one ends up with

$$\widehat{\Pi}(h)=\left[(\beta_4,\gamma_4)\alpha_4(\beta_3,\gamma_3)\alpha_3(\beta_2,\gamma_2)\alpha_2(\beta_1,\gamma_1)\alpha_1\right]. \tag{3.16}$$

Notice that, in all cases, we have one free parameter. Nevertheless, it is not always possible to get solutions with all $\alpha_j>0$, $\beta_j>0$, in particular for kernel (3.14). In fact, in that case there are no solutions with all $\alpha_j>0$ for alternative (a), whereas for (b) and (c) one of the β_j has to be necessarily negative.

For kernel (3.15) we have, by contrast, 1-parameter families of solutions involving only positive coefficients. Particular choices are collected in Table 4. We recall that in this case the overall method for N integration steps reads

$$\begin{aligned}\widetilde{\mathcal{S}}_N(h)=&\left[\alpha_1(\beta_1,\gamma_1)\alpha_2(\beta_2,\gamma_2)\alpha_3(\beta_3,\gamma_3)\alpha_4(\beta_4,\gamma_4)\right]\cdot(\mathcal{S}_{\rm BAB}(h))^{N-2}\\ &\cdot\left[(\beta_4,\gamma_4)\alpha_4(\beta_3,\gamma_3)\alpha_3(\beta_2,\gamma_2)\alpha_2(\beta_1,\gamma_1)\alpha_1\right].\end{aligned}$$

4 Numerical comparisons

In this section we compare the new 4th-order modified processed splitting methods with respect to the most efficient schemes also involving modified potentials and positive coefficients we have found in the literature.

4.1 Imaginary time propagation in 1-d

As a first illustration we apply the previous methods to the numerical integration of the one-dimensional Schrödinger equation in imaginary time for three different potentials: the harmonic oscillator, a double well such that the minima of the potential can be approximated by cubic functions, and finally a potential where higher derivatives are relevant near the minimum.

As stated in the introduction, in this diffusive problem the solution converges to the ground state which is mainly concentrated around the minimum of the potential. We measure the error in the normalized solution at the final time as

$$E_r\equiv\left\|\frac{u(\tau_f)}{\|u(\tau_f)\|}-\frac{u_{app}(\tau_f)}{\|u_{app}(\tau_f)\|}\right\|$$

and depict this error as a function of the computational cost measured as the number of fast Fourier transforms (FFTs) and inverses of FFTs computed.

First we analyze the performance of the best 4th-order methods from the literature with modified potentials. Next, we analyze the performance of the 1-stage and the new 2-stage kernels with processors that are obtained applying different optimization criteria. Finally, we collect the best processed and non-processed methods to illustrate their relative performance taking the Strang method as a reference. Specifically, we test the following integrators:

Table 4: Coefficients of processed splitting methods with kernel of type BAB with 2 stages and all positive coefficients. The different processors are chosen according with criteria (a), (b) and (c).

Type	Kernel	Starter	
		$\alpha_1 = 0.085901112008607587690928693459$	(a)
		$\beta_1 = 0.212698346714285489082694412338$	
		$\gamma_1 = 1/20000$	
		$\alpha_2 = 0.344269607777567701053555680439$	
		$\beta_2 = 0.477571645650134700450689457043$	
		$\gamma_2 = 0.006356334205889079278610714346 4$	
		$\alpha_3 = 0.499160597130554034892205778 69$	
		$\beta_3 = 0.107615382576486586889760168322$	
		$\gamma_3 = 0.001520954618870958598557362734 54$	
		$\alpha_4 = 0.070668683083270676363309847409$	
		$\beta_4 = 0.202114625059093223576855962 30$	
		$\gamma_4 = 0.000439633064716702370928763003143$	
		$\alpha_1 = 0.081400719358833550685242298084$	(b)
		$\beta_1 = 0.213974166240535023746064422594$	
	$b_1 = \frac{27}{100}$	$\gamma_1 = 0$	
	$c_1 = \frac{1597}{1440000}$	$\alpha_2 = 0.359973312089188663628848658435$	
BAB	$a_2 = \frac{1}{2}$	$\beta_2 = 0.489218997657784225611758847163$	
	$b_2 = \frac{23}{50}$	$\gamma_2 = 0.006383941828680749399669 0114472$	
	$c_2 = \frac{6047}{720000}$	$\alpha_3 = 0.533271252488022531567730572505$	
		$\beta_3 = 0.170371266269132266613375096529$	
		$\gamma_3 = 0.010246795170879377422282291 8119$	
		$\alpha_4 = 0.025354716063955254118178470 9748$	
		$\beta_4 = 0.126435569832548484028801633 71$	
		$\gamma_4 = -0.007702960372378025636599968 6550$	
		$\alpha_1 = 0.085835304465630711824262626400$	(c)
		$\beta_1 = 0.217482775489438674951915790962$	
		$\gamma_1 = 1/10000$	
		$\alpha_2 = 0.353863857161384364666360893974$	
		$\beta_2 = 0.482376576131251183368714037601$	
		$\gamma_2 = 0.006221170395362736619593811 9902$	
		$\alpha_3 = 0.531192662483216027224466133435$	
		$\beta_3 = 0.174369958075879961757677904504$	
		$\gamma_3 = 0.007262693793548439673917370 9272$	
		$\alpha_4 = 0.029108175889768896284910346 1909$	
		$\beta_4 = 0.125770690303430179921692266 93$	
		$\gamma_4 = -0.004835274510931325703953715661 79$	

- $S_1^{[2,a]}$: the Strang splitting of type ABA, used here as a reference method;
- $Sm_2^{[4,a]}$, $Sm_2^{[4,b]}$: the 2-stage ABA and BAB methods (2.15) and (2.14), with positive coefficients and modified potentials;
- $Sm_3^{[4,a]}$, $Sm_3^{[4,b]}$: the most efficient 3-stage methods of type ABA and BAB proposed in [24] and collected in (2.16);
- $Sm_4^{[4,a]}$, $Sm_4^{[4,b]}$: the most efficient 4-stage methods of type ABA and BAB proposed in [24] and collected in (2.17);
- $SP_1^{[4]}$: the modified processed scheme with a 1-stage kernel of type ABA with symmetric processor and positive coefficients (3.13);
- $SP_{2,\alpha}^{[4]}$: the 2-stage BAB methods with symmetric processor built according to criterion (a), (b) or (c) ($\alpha = a,b,c$) and positive coefficients, as collected in Table 4.

Notice that the previous notation indicates the order of the scheme (superscript), the number of stages (subscript), the particular composition type (a: ABA, b: BAB), the presence of modified potentials without processor (m) and the presence of modified potentials with processor (P).

Harmonic oscillator potential. We first take

$$\hat{V}(x) = \frac{1}{2}x^2$$

and the initial wave function $\psi_0(x) = \sigma|\cos(x)|e^{-(x-2)^2}$, where σ is the normalizing constant. Since the solution decays rapidly, we can safely truncate the infinite spatial domain to the periodic interval $[-L,L]$, provided L is sufficiently large. Specifically, we take $L=10$ and set up a uniform grid $x_1,\cdots,x_N$ in the interval, with $N=128$, and integrate until the final time $\tau_f = 2$. Notice that for this problem the error terms $E_{5,1} = E_{5,2} = 0$, so that the error is dominated by $f_{5,3}$ and $f_{5,4}$.

Fig. 1 contains three diagrams collecting, in a log-log scale, the error E_r vs. the number of FFTs for the different integrators. Specifically, the top left diagram shows the results achieved by the 4th-order conventional methods of type ABA and BAB with 2, 3 and 4 stages, modified potentials and positive coefficients. We observe an improvement in the accuracy with the number of stages per step (number of free parameters in the scheme) at the cost of an slightly worst performance at low accuracies. Among them, we choose the 4-stage BAB scheme as the most efficient one. The top right panel collects the results obtained by the new processed methods. Since all methods using the 2-stage kernel have been designed so that $f_{5,3} = f_{5,4} = 0$, then they achieve in practice order 6. For this problem, all three methods with the 2-stage kernel show a very similar performance. At low accuracies (low number of FFTs) the cost of the pre- and post-processor is not negligible. Finally, in the bottom diagram we compare the most efficient schemes from the

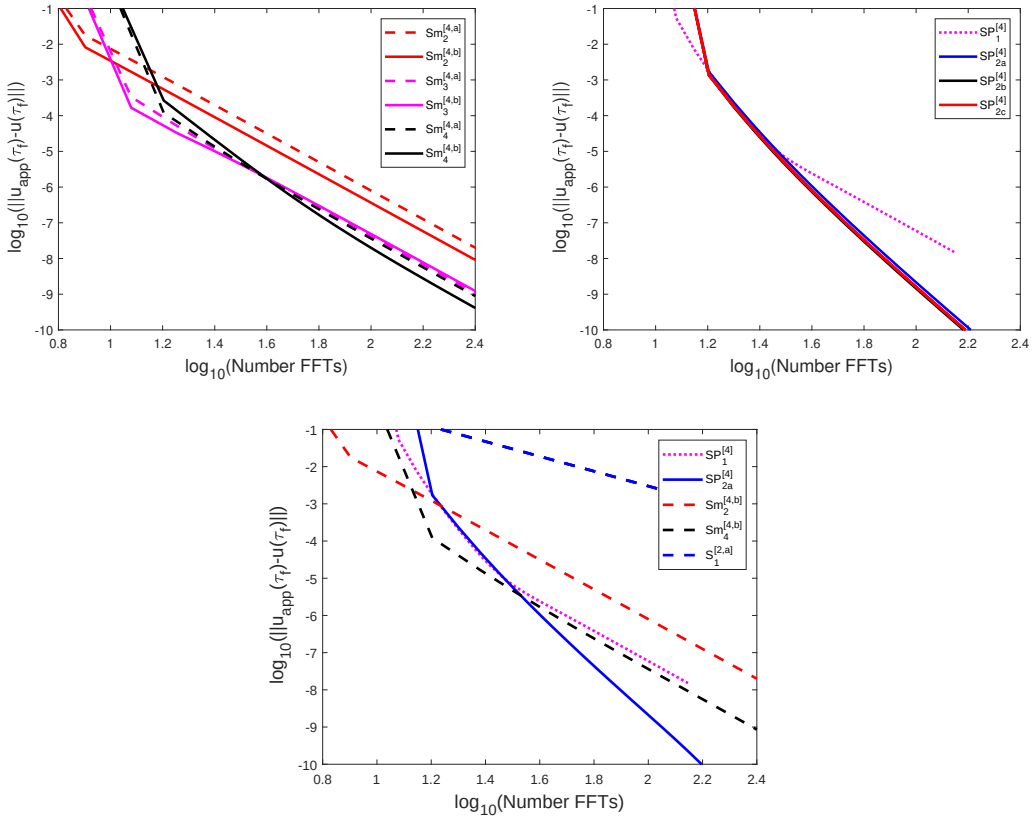

Figure 1: Harmonic oscillator potential. Top left: the 4th-order non-processed methods for the 2, 3 and 4-stage ABA and BAB methods with modified potentials; Top right: the 4th-order 1-stage and 2-stage processed methods; Bottom figure: the best methods among the previous ones jointly with the Strang method (used as a reference method).

previous graphs together with the results obtained by the Strang method. We observe an improvement of the new processed 2-stage kernel scheme with respect to the most efficient conventional integrators involving modified potentials at high accuracies. As mentioned, the performance at low accuracies is reduced due to the cost of the processor (this cost would not be considered in problems where e.g. one is only interested in the trace of the transition matrix).

Double well potential. The same set of experiments is carried out for the quartic oscillator

$$\hat{V}(x)=10-\frac{1}{2}x^2+\frac{1}{160}x^4=\frac{1}{160}(x^2-40)^2$$

with initial condition $\psi_0(x)=\sigma|\cos(x)|e^{-(x-6)^2}$. Here again σ is the normalizing constant and the initial wave function is allocated near one of the minima of the potential. In this case we take as the spatial interval the periodic domain $[-13,13]$ and $N=128$ grid points

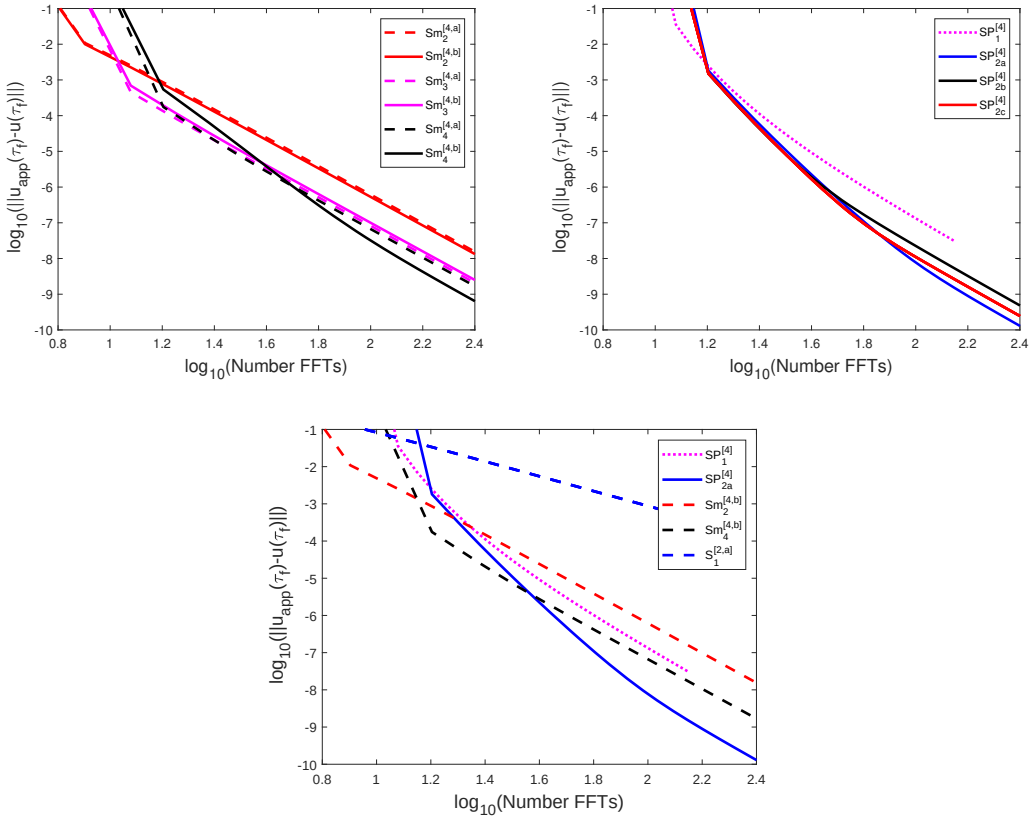

Figure 2: Same as Fig. 1 but for the double well potential.

for the Fourier semidiscretization. The final integration time is $\tau_f = 2$. The results are collected in Fig. 2, where the same pattern in the top left diagram can be observed. In the top right diagram we see, as expected, a small improvement by the scheme $\mathrm{SP}^{[4]}_{2,a}$ which would correspond to a sixth-order method for a cubic potential (this is essentially the case near the minimum of the potential). In the bottom diagram we compare the most efficient schemes from the previous graphs, where the superiority of the new method is visible for high accuracy. As in the previous case, at low accuracies the relative cost of the pre- and post-processors is significant.

Pöschl–Teller potential. As a third illustration in the context of the ITP algorithm we take the potential

$$\hat{V}(x) = \frac{\lambda(\lambda+1)}{2}(1-\mathrm{sech}(x)^2)$$

with $\lambda = 5$ and initial condition $\psi_0(x) = \sigma|\cos(x)|\mathrm{e}^{-x^2}$. Our spatial interval is $[-10,10]$, the uniform grid has $N = 128$ points and the final time is $\tau_f = 2$. Fig. 3 shows the results from the methods which where previously chosen as the most efficient ones. In this case,

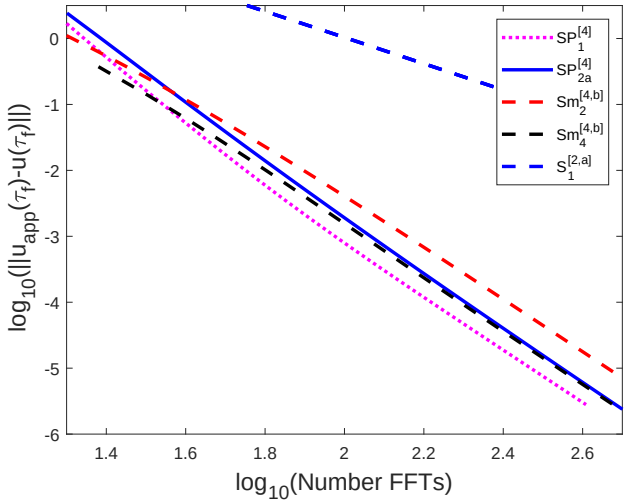

Figure 3: Same as Fig. 1 but for the Pöschl–Teller potential.

all choices of the processor for the 2-stage kernel lead essentially to the same results, but now the 1-stage kernel shows the best performance.

It is important to remark the superiority of all the methods with modified potentials with respect to the frequently used second order Strang method and that this superiority increases with the desired accuracy. The 4-stage BAB method $\mathrm{Sm}_4^{[4,b]}$ shows, in general, a better performance than the frequently used 2-stage BAB method $\mathrm{Sm}_2^{[4,b]}$ and the new processed methods are even superior when high accuracy is desired (and also at low accuracies if the cost of the pre- and post-processor can be neglected).

4.2 1-d, 2-d and 3d linear evolution problems

As further test problems, we consider linear parabolic equations of the form

$$\frac{\partial}{\partial t}u(x,t)=\tfrac{1}{2}\Delta u(x,t)+\hat{V}(x)u(x,t),\quad t\in[0,\tau_f],\quad u(x,0)=u_0(x)$$

in one, two and three dimensions, to study how the relative performance of the methods change with the dimension. On the one hand, we prescribe periodic problem data for $d=1,2,3$

$$u_0(x)=u_0(x_1,\cdots,x_d)=c\prod_{j=1}^{d}\sin^2\left(\tfrac{x_j+a}{a}\pi\right),$$

$$\hat{V}(x)=c\prod_{j=1}^{d}\left(1-\cos^2\left(\tfrac{x_j+a}{2a}\pi\right)\right),$$

$$x\in[-L,L]^d,\quad L=10,\quad c=10,\quad d=1,2,3,\quad \tau_f=10.$$

On the other hand, we study the time evolution of a localized initial state under a standard quadratic potential

$$u_0(x)=\mathrm{e}^{-5\|x\|^2},\quad V(x)=-\|x\|^2,\quad x\in[-10,10]^d,\quad d=1,2,3,\quad \tau_f=1.$$

We take a uniform grid $x_{j,1},\cdots,x_{j,N}$, $j=1,\cdots,d$, with $N=512$ for $d=1$ leading to a system of dimension $M=512$, $N=128$ for $d=2$ leading to a system of dimension $M=128^2=16384$, and finally with $N=64$ for $d=3$, so $M=64^3=262144$.

The time integration is performed with the methods which were previously selected as the most efficient, and also include for comparison the classical second-order Strang splitting. In Fig. 4, and for simplicity, we denote them as follows:

- $\mathrm{S}_1^{[2,a]}$, referred to as Order 2 (Strang);
- $\mathrm{Sm}_2^{[4,b]}$, called here Order 4 (Chin);
- $\mathrm{Sm}_4^{[4,b]}$, referred to as Order 4 (Omelyan);
- $\mathrm{SP}_1^{[4]}$, denoted by Order 4 (Rowland); and
- $\mathrm{SP}_{2a}^{[4]}$, called Order 4 (Novel).

With regard to the spatial discretization, we apply as before the Fourier pseudo-spectral method on an equidistant mesh and measure the computational effort by the total number of fast Fourier transforms and inverse fast Fourier transforms. In order to determine the global errors at the final time, a numerical reference solution is computed by dividing the smallest time stepsize in half. The obtained results, given in Fig. 4, are consistent with our former observations and confirm the high accuracy of the novel scheme $\mathrm{SP}_{2a}^{[4]}$ at a reasonable computational cost in different settings.

5 Conclusions

We have presented a novel class of splitting methods for numerically solving linear parabolic problems that appear in many physical problems, such as the imaginary time propagation of the Schrödinger equation. Splitting methods are especially useful in this context, due to their simple structure and good performance. For periodic problems or when the solution vanishes far from the region of interest, the order reduction of the methods usually does not occur and higher order schemes can be of interest. Splitting methods of order greater than two necessarily have negative coefficients and thus can not be used to propagate the unbounded operator associated to the Laplacian. This problem can be partially solved by incorporating into the scheme some appropriated modified potentials, thus leading to fourth-order integrators with positive coefficients. In fact, order four constitutes the upper limit for this family of methods.

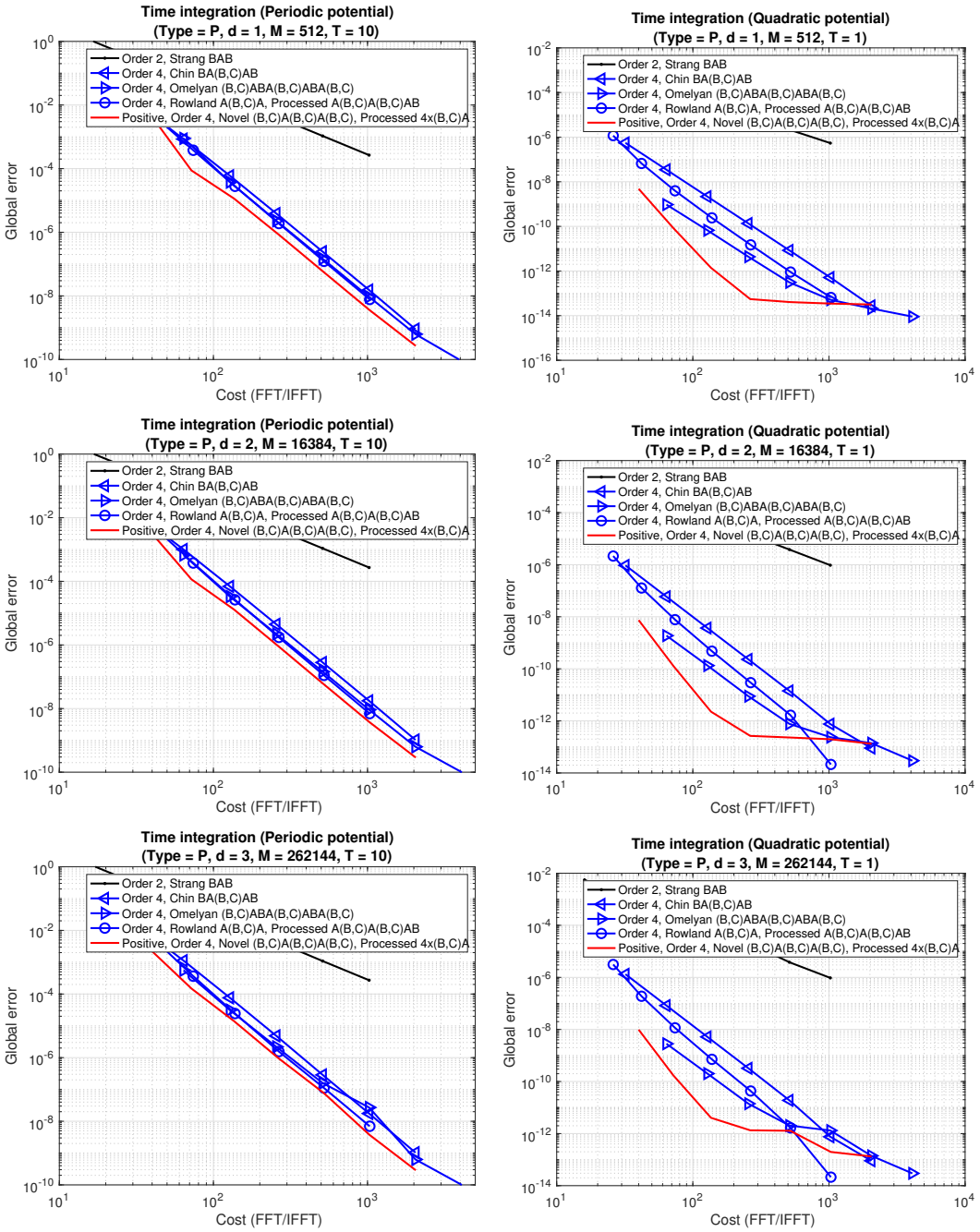

Figure 4: Time integration of a linear evolution equation of parabolic type involving a periodic potential (left) or a standard quadratic potential (right). The Fourier pseudo-spectral space discretization in dimension d is based on a total number of M equidistant grid points. For the classical Strang splitting method and different fourth-order schemes, we depict the global errors at final time τ_f versus the computational cost, measured by the number of FFTs and their inverse.

We have carried out a search in the literature of methods of order four that incorporate modified potentials in their formulation and have positive coefficients. We have observed the limitations to improve their performance by applying the standard strategy of augmenting the number of stages to reduce the error, and we have proposed a generalization of the processing technique to achieve higher accuracy and better stability with a reduced computational cost. Methods with processors, in general, have negative coefficients in the pre- and post-processors due to consistency and also because one has to compute a scheme to start and its inverse to conclude the integration. Both drawbacks are resolved by considering symmetric processors with a starter, so that new families of fourth-order integrators with modified potentials and positive coefficients can be generated. An error analysis is carried indicating that the new methods possess smaller errors. We thus end up with new schemes with a similar complexity as previous splitting methods with modified potentials, but with a superior efficiency, as shown by the numerical examples collected here.

The new proposed methods can also be safely applied in any situation where the schemes of [12] and [24] are used, and in particular to the numerical integration of the Schrödinger equation in real time. Since they have small and positive coefficients, one can expect to be highly efficient in situations where low to medium accuracies are required (usually, higher order methods are preferable when very high accuracy is desired).

The generalization of the proposed methods to nonlinear both parabolic and Schrödinger equations is one of our objective in current research investigation. In [18] it is proven that methods with modified potentials retain their classical order of accuracy provided the solution is sufficiently regular. In the case of low regularity problems an order reduction is expected and different techniques should be used (see e.g. [20]).

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