Compact schemes for laser–matter interaction in Schrödinger equation

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Abstract

Numerical solutions for laser-matter interaction in Schrödinger equation has many applications in theoretical chemistry, quantum physics and condensed matter physics. In this paper we introduce a methodology which allows, with a small cost, to extend any fourth-order scheme for Schrödinger equation with time-independent potential to a fourth-order method for Schrödinger equation with laser potential. These fourth-order methods improve upon many leading schemes of order six due to their low costs and small error constants.

1 Introduction

In this paper we are concerned with developing highly efficient numerical approaches for laser-matter interaction in the Schrödinger equation,

$$\mathrm{i}\varepsilon\partial_t u(\boldsymbol{x},t) = \big[-\varepsilon^2\Delta + V_0(\boldsymbol{x}) + \boldsymbol{e}(t)^\top\boldsymbol{x}\big]u(\boldsymbol{x},t),\ u(\boldsymbol{x},0) = u_0(\boldsymbol{x}), \tag{1.1}$$

where $t \geq 0$, $\boldsymbol{x} = (x_1,\ldots,x_n) \in \mathbb{R}^n$ and the laser term $\boldsymbol{e}(t) = (e_1(t),\ldots,e_n(t))$ is an $\mathbb{R}^n$ valued function of t. If the direction of the laser is assumed to be fixed, $\boldsymbol{e}(t)$ is of the form

$$\boldsymbol{e}(t) = e(t)\hat{\boldsymbol{\mu}}, \quad e(t) \in \mathbb{R},\ \hat{\boldsymbol{\mu}} \in \mathbb{R}^n,\ \|\hat{\boldsymbol{\mu}}\|_2 = 1,$$

A very specific case, which is frequently used is $\hat{\boldsymbol{\mu}} = (1,0,\ldots,0)$, so that

$$V(\boldsymbol{x},t) = V_0(\boldsymbol{x}) - e(t)x_1.$$

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The parameter ε in (1.1) acts like Planck's constant. This parameter is 1 when working in the atomic scaling and is very small, $0 < \varepsilon \ll 1$, when working in the semiclassical regime. The methods developed in this paper are equally effective in both regimes and will cover the special cases of lasers as well.

Schrödinger equations under the influence of lasers play a significant role in quantum physics and theoretical chemistry, as they aid in the simulation and design of systems and processes at atomic and molecular scales. Highly accurate and cost effective schemes are required, for instance, in applications such as optimal control strategies for shaping lasers where the numerical solutions for these equations are used repeatedly within an optimisation routine (Amstrup, Doll, Sauerbrey, Szabó & Lorincz 1993, Meyer, Wang & May 2006, Coudert 2018).

The presence of time-dependent potentials in the Schrödinger equation makes the challenge of designing an efficient method significantly harder than the case of time-independent potentials since typical strategies for this case require utilisation of Magnus expansion at each time step, which involves nested integrals of nested commutators of large matrices.

The design of effective methods for time-dependent potentials is, therefore, a very challenging and active research area in theoretical chemistry, quantum physics and numerical mathematics. A wide range of methodologies has been developed to effectively handle this case (Tal-Ezer, Kosloff & Cerjan 1992, Peskin, Kosloff & Moiseyev 1994, Sanz-Serna & Portillo 1996, Tremblay & Carrington Jr. 2004, Klaiber, Dimitrovski & Briggs 2009, Ndong, Tal-Ezer, Kosloff & Koch 2010, Alvermann & Fehske 2011, Blanes, Casas & Murua 2017*a*, Blanes, Casas & Thalhammer 2017*b*, Schaefer, Tal-Ezer & Kosloff 2017, Iserles, Kropielnicka & Singh 2018*a*, Iserles, Kropielnicka & Singh 2018*b*). The aforementioned methods can indeed be applied to (1.1). However, apart the third-order method from (Klaiber et al. 2009), they are not specialised for the case of lasers, where the structure of the potential can be exploited to yield more efficient methods.

In this paper we propose a fourth-order numerical method, highly specialised for the Schrödinger equation under the influence of a laser (1.1). The main merits of the proposed method are its low costs and the ease with which existing fourth-order implementations for the time-independent potentials can be extended to handle time-dependent potentials. The cost of the proposed methods is only marginally higher than the fastest fourth-order methods dedicated for Schrödinger equation with time-independent potentials, $V_0(\boldsymbol{x})$.

The two main ingredients in the derivation of our method are (i) the simplification of commutators in the Magnus expansion by exploiting the special form of laser potentials (ii) approximating the exponential of this Magnus expansion by appropriate fourth-order splittings, again exploiting the special form of the potential. In step (ii), we face a choice – we may opt for (a) a combination of Strang splitting with a fourth-order scheme for time-independent potentials or (b) a compact splitting featuring positive coefficients directly for this Magnus expansion.

In the application of Strang splitting in the first case, we extract the smallest component of the Magnus expansion as the outer term (with a cost of only two additional FFTs, due to its special structure). This yields fourth order accuracy. The inner term can then be approximated with any of the many suitable methods for Schrödinger equation with time-independent potentials. This may be the highly optimised splitting of (Blanes & Moan 2002), the compact splitting of (Chin & Chen 2002) (if the gradient of potential is available) or any other fourth-order method, depending on the requirements or availability of an existing implementation.

In Section 2 we obtain, via simplification of commutators, the optimal form of Magnus expansion, which can be handled efficiently. In Section 3 we present application of Strang splitting, explaining why it serves here as a fourth order method. In its subsections, in turn, we describe two (out of many) options that could be applied for the exponentiation of the inner exponential. We also briefly discuss the computational aspects and explain the low cost of this approach. Section 4 pursues the second alternative, presenting a compact splitting scheme applied directly to the Magnus expansion of Section 2. Numerical examples are described in Section 5, while our conclusions are summarised in Section 6.

2 Application of the Magnus expansion

The Schrödinger equation (1.1) can be rewritten in the form

$$\partial_t u(\boldsymbol{x},t) = \mathcal{A}(\boldsymbol{x},t)u(\boldsymbol{x},t), \quad \boldsymbol{x}\in\mathbb{R}^n,\ t\ge 0,\ u(\boldsymbol{x},0)=u_0(\boldsymbol{x}), \tag{2.1}$$

where $\mathcal{A}(\boldsymbol{x},t) = \mathrm{i}\varepsilon\Delta - \mathrm{i}\varepsilon^{-1}\left(V_0(\boldsymbol{x}) + \boldsymbol{e}(t)^\top\boldsymbol{x}\right)$. A fourth-order numerical scheme for this equation can be obtained by resorting to a Magnus expansion (Magnus 1954),

$$\boldsymbol{u}(t+h) = \mathrm{e}^{\Theta_2(t+h,t)}\boldsymbol{u}(t),$$

where the Magnus series has been truncated to the first two terms[1],

$$\Theta_2(t+h,t) = \int_0^h \mathcal{A}(t+\zeta)\,\mathrm{d}\zeta - \tfrac{1}{2}\int_0^h\int_0^\zeta [\mathcal{A}(t+\xi), A(t+\zeta)]\,\mathrm{d}\xi\,\mathrm{d}\zeta.$$

It is a simple matter to verify that

$$\begin{aligned}\Theta_2(t+h,t) = {}& \mathrm{i}h\varepsilon\Delta - \mathrm{i}h\varepsilon^{-1}V_0(\boldsymbol{x}) - \mathrm{i}\varepsilon^{-1}\left(\int_0^h \boldsymbol{e}(t+\zeta)\,\mathrm{d}\zeta\right)^\top \boldsymbol{x}\\ & -\tfrac{1}{2}\left(\int_0^h\int_0^\zeta [\boldsymbol{e}(t+\zeta)-\boldsymbol{e}(t+\xi)]\,\mathrm{d}\xi\,\mathrm{d}\zeta\right)^\top [\Delta,\boldsymbol{x}].\end{aligned}$$

We note that

$$[\Delta,\boldsymbol{x}]u = \sum_{j=1}^n \left(\partial^2_{x_j}(\boldsymbol{x}u) - \boldsymbol{x}\partial^2_{x_j}u\right) = 2\nabla u,$$

and the fact that the integral over the triangle in Θ_2 can be rewritten as a univariate integral,

$$\int_0^h\int_0^\zeta [\boldsymbol{e}(t+\zeta)-\boldsymbol{e}(t+\xi)]\,\mathrm{d}\xi\,\mathrm{d}\zeta = 2\int_0^h \left(\zeta - \tfrac{h}{2}\right)\boldsymbol{e}(t+\zeta)\,\mathrm{d}\zeta.$$

[1] Commutator of the operators $\mathcal{A}$ and $\mathcal{B}$ is defined here as $[\mathcal{A},\mathcal{B}] = \mathcal{A}\circ\mathcal{B} - \mathcal{B}\circ\mathcal{A}$.

Using these relations, we recast the Magnus expansion up to order four in the form

$$\Theta_2(t+h,t) = \mathrm{i}h\varepsilon\Delta - \mathrm{i}h\varepsilon^{-1}\widetilde{V}(\boldsymbol{x},t,h) - \boldsymbol{s}(t,h)^\top\nabla, \tag{2.2}$$

where

$$\widetilde{V}(\boldsymbol{x},t,h) = V_0(\boldsymbol{x}) + \boldsymbol{r}(t,h)^\top\boldsymbol{x}, \tag{2.3}$$

$$\boldsymbol{r}(t,h) = \tfrac{1}{h}\int_0^h \boldsymbol{e}(t+\zeta)\,\mathrm{d}\zeta, \tag{2.4}$$

$$\boldsymbol{s}(t,h) = 2\int_0^h \left(\zeta - \tfrac{h}{2}\right)\boldsymbol{e}(t+\zeta)\,\mathrm{d}\zeta. \tag{2.5}$$

The numerical exponentiation of (2.2) is incredibly costly unless it is split properly. In Sections 3 and 4, we will explore two fourth-order exponential splitting strategies for approximating this exponential.

3 A Strang splitting of the Magnus expansion

A Strang splitting of $\Theta_2(t+h,t) = X+Y$, with $X = \mathrm{i}h\varepsilon\Delta - \mathrm{i}h\varepsilon^{-1}\widetilde{V}(\boldsymbol{x},t,h)$ and $Y = -\boldsymbol{s}(t,h)^\top\nabla$,

$$\exp\left(-\tfrac{1}{2}\boldsymbol{s}(t,h)^\top\nabla\right)\mathrm{e}^{\mathrm{i}h\varepsilon\Delta - \mathrm{i}h\varepsilon^{-1}\widetilde{V}(\boldsymbol{x},t,h)}\exp\left(-\tfrac{1}{2}\boldsymbol{s}(t,h)^\top\nabla\right), \tag{3.1}$$

is an order-four method, not order two as usually expected of Strang splitting.

To see this, note that according to the symmetric Baker–Campbell–Hausdorff (sBCH) formula,

$$\mathrm{e}^{\frac{1}{2}Y}\mathrm{e}^{X}\mathrm{e}^{\frac{1}{2}Y} = \mathrm{e}^{\mathrm{sBCH}(Y,X)},\ \ \mathrm{sBCH(X,Y)} = X+Y-\left(\tfrac{1}{24}[[Y,X],X] + \tfrac{1}{12}[[Y,X],Y]\right)+\text{h.o.t.},$$

the Strang splitting approximates the exponential of $X+Y$ up to the commutators $\frac{1}{24}[[Y,X],X]+\frac{1}{12}[[Y,X],Y]$. However, unlike the usual application of Strang splitting, where $X,Y = \mathcal{O}(h)$, the second term in the Magnus expansion, $Y = -\boldsymbol{s}(t,h)^\top\nabla$, scales as $\mathcal{O}\left(h^3\right)$ (Iserles, Munthe-Kaas, Nørsett & Zanna 2000). Consequently, the largest commutator in our case is $\frac{1}{24}[[Y,X],X] = \mathcal{O}\left(h^5\right)$, and the error in the application of Strang splitting used for deriving (3.1) is $\mathcal{O}\left(h^5\right)$.

The central exponent in (3.1) can now be approximated via any order-four method for time-independent potentials. In other words, as long as an effective integrator for time-independent potentials is available, it need only be modified in the following manner in order to convert it to an order-four scheme for time-dependent potentials:

1. In each step $\widetilde{V}(\boldsymbol{x},t,h)$ has to be recomputed. However, as we can see from (2.3), this only requires the computation of the time-integral of the laser pulse (2.4), and not the re-evaluation of V_0, which needs to be computed only once.

2. The potential $\widetilde{V}$ should be used in place of V_0 in the existing fourth-order scheme for time-independent potentials.

3. Lastly, we need to compute the outermost exponentials, $\exp\left(-\frac{1}{2}\boldsymbol{s}(t,h)^\top\nabla\right)$. Note that this can be combined across two consecutive steps of (3.1),

$$\exp\left(-\tfrac{1}{2}\boldsymbol{s}(t+h,h)^\top\nabla\right)\exp\left(-\tfrac{1}{2}\boldsymbol{s}(t,h)^\top\nabla\right) = \exp\left(-\tfrac{1}{2}\left[\boldsymbol{s}(t+h,h)+\boldsymbol{s}(t,h)\right]^\top\nabla\right),$$

   so that the overall scheme only involves one additional exponential per step.

In general, the evaluation of the extra exponential should be no more expensive than the exponential of the Laplacian, which is routinely employed in schemes for the Schrödinger equation with time-independent potentials. In some cases it is possible to combine this exponential in such a manner that no additional cost is incurred in the scheme, as we shall see in Section 3.1.3.

In the following sections we will consider two concrete examples of order-four methods for time-independent potentials for approximation of the central exponential of (3.1).

3.1 Approximation of the inner term

A wide range of exponential splittings are readily available and easily implementable for the approximation of the Schrödinger equation with time-independent potentials (McLachlan & Quispel 2002, Blanes, Casas & Murua 2006, Blanes, Casas & Murua 2008).

The choice of appropriate exponential splittings may be governed by a need for fewer exponentials, lower error constants or other constraints such as positivity of coefficients. The examples we will consider are the highly optimised order-four exponential splitting of (Blanes & Moan 2002) and the compact splitting scheme from (Chin & Chen 2002).

3.1.1 Classical splittings

The optimised splitting from (Blanes & Moan 2002) for $X, Y = \mathcal{O}(h)$,

$$\mathrm{e}^{X+Y} = \mathrm{e}^{a_1X}\mathrm{e}^{b_1Y}\mathrm{e}^{a_2X}\mathrm{e}^{b_2Y}\mathrm{e}^{a_3X}\mathrm{e}^{b_3Y}\mathrm{e}^{a_4X}\mathrm{e}^{b_3Y}\mathrm{e}^{a_3X}\mathrm{e}^{b_2Y}\mathrm{e}^{a_2X}\mathrm{e}^{b_1Y}\mathrm{e}^{a_1X} + \mathcal{O}\left(h^5\right), \quad (3.2)$$

where

$$\begin{aligned} a_1 &= 0.0792036964311957, & b_1 &= 0.209515106613362,\\ a_2 &= 0.353172906049774, & b_2 &= -0.143851773179818,\\ a_3 &= -0.0420650803577195, & b_3 &= 1/2 - b_1 - b_2,\\ a_4 &= 1 - 2(a_1+a_2+a_3), \end{aligned}$$

is known to have a very small error constant (McLachlan & Quispel 2002). The choices

$$X = \mathrm{i}h\varepsilon\Delta = \mathcal{O}(h), \quad Y = -\mathrm{i}h\varepsilon^{-1}\widetilde{V}(\boldsymbol{x},t,h) = \mathcal{O}(h),$$

result in an order-four splitting for the central exponent of (3.1). Combining this with the outer exponents in (3.1) completes one example of a fourth-order scheme for time-dependent potentials.

3.1.2 Compact splittings

Another concrete example results from using (Chin & Chen 2002) for the approximation of the central exponential,

$$\mathrm{e}^{X+Y} = \mathrm{e}^{\frac{1}{6}Y}\mathrm{e}^{\frac{1}{2}X}\mathrm{e}^{\frac{2}{3}(Y+\frac{1}{48}[[X,Y],Y])}\mathrm{e}^{\frac{1}{2}X}\mathrm{e}^{\frac{1}{6}Y} + \mathcal{O}\left(h^5\right). \tag{3.3}$$

In the case of

$$X = \mathrm{i}h\varepsilon\Delta = \mathcal{O}\left(h\right), \quad Y = -\mathrm{i}h\varepsilon^{-1}\widetilde{V}(\boldsymbol{x},t,h) = \mathcal{O}\left(h\right),$$

while the nested commutator reduces to a function,

$$[[X,Y],Y] = -\mathrm{i}h^3\varepsilon^{-1}[[\Delta,\widetilde{V}],\widetilde{V}] = -2\mathrm{i}h^3\varepsilon^{-1}\sum_{j=0}^{n}[(\partial_{x_j}\widetilde{V})\partial_{x_j},\widetilde{V}] = -2\mathrm{i}h^3\varepsilon^{-1}\nabla\widetilde{V}^\top\nabla\widetilde{V}.$$

Thus, (3.3) reduces to

$$\mathrm{e}^{-\frac{1}{6}\mathrm{i}h\varepsilon^{-1}\widetilde{V}}\mathrm{e}^{\frac{1}{2}\mathrm{i}h\varepsilon\Delta}\mathrm{e}^{-\frac{2}{3}\mathrm{i}h\widehat{V}}\mathrm{e}^{\frac{1}{2}\mathrm{i}h\varepsilon\Delta}\mathrm{e}^{-\frac{1}{6}\mathrm{i}h\varepsilon^{-1}\widetilde{V}}, \tag{3.4}$$

where $\widehat{V}$ is an $\mathcal{O}\left(h^3\right)$ perturbation of $\widetilde{V}$,

$$\widehat{V} = \widetilde{V} - \tfrac{h^2}{24}(\nabla\widetilde{V})^\top(\nabla\widetilde{V}). \tag{3.5}$$

The overall order-four scheme is obtained by substituting the central exponent in (3.1) with (3.4),

$$\mathrm{e}^{-\frac{1}{2}\boldsymbol{s}(t,h)^\top\nabla}\mathrm{e}^{-\frac{1}{6}\mathrm{i}h\varepsilon^{-1}\widetilde{V}}\mathrm{e}^{\frac{1}{2}\mathrm{i}h\varepsilon\Delta}\mathrm{e}^{-\frac{2}{3}\mathrm{i}h\varepsilon^{-1}\widehat{V}}\mathrm{e}^{\frac{1}{2}\mathrm{i}h\varepsilon\Delta}\mathrm{e}^{-\frac{1}{6}\mathrm{i}h\varepsilon^{-1}\widetilde{V}}\mathrm{e}^{-\frac{1}{2}\boldsymbol{s}(t,h)^\top\nabla}. \tag{3.6}$$

This scheme involves the evaluation of $\nabla\widetilde{V}$. However, the typically expensive part, ∇V_0, needs to be computed only once since

$$\nabla\widetilde{V} = \nabla V_0 + \boldsymbol{r}(t,h).$$

3.1.3 Combining exponentials

A further optimisation is possible in some cases. Upon replacing the inner exponential in (3.1) by (3.2) in Section 3.1.1, it should be observed that the outermost exponentials commute and, therefore, can be combined,

$$\exp\left(-\tfrac{1}{2}\boldsymbol{s}(t,h)^\top\nabla\right)\mathrm{e}^{\mathrm{i}a_1h\varepsilon\Delta} = \exp\left(\mathrm{i}a_1h\varepsilon\Delta - \tfrac{1}{2}\boldsymbol{s}(t,h)^\top\nabla\right).$$

In practice, this combined exponential is often not much harder to compute than the exponential of the Laplacian.

For instance, under spectral collocation the differentiation matrices are circulant and are diagonalised via Fourier transforms[2],

$$\partial_x^k \rightsquigarrow \boldsymbol{D}_{k,x} = \mathcal{F}_x^{-1}\mathcal{D}_{c_{k,x}}\mathcal{F}_x,$$

[2]We use $\rightsquigarrow$ to denote discretisation.

where $\mathcal{D}_{c_{k,x}}$ is a diagonal matrix and the values along its diagonal, $c_{k,x}$, comprise the symbol of the kth differentiation matrix, $\boldsymbol{D}_{k,x}$. In two dimensions the exponential of the Laplacian term, $\mathrm{i}a_1 h\varepsilon\Delta$, alone is implemented as

$$\mathrm{e}^{\mathrm{i}a_1 h\varepsilon\Delta} v \rightsquigarrow \mathcal{F}_x^{-1}\mathcal{D}_{\exp(\mathrm{i}a_1 h\varepsilon c_{2,x})}\mathcal{F}_x\mathcal{F}_y^{-1}\mathcal{D}_{\exp(\mathrm{i}a_1 h\varepsilon c_{2,y})}\mathcal{F}_y\boldsymbol{v},$$

using four Fast Fourier Transforms (FFTs). Similarly,

$$\mathrm{e}^{\mathrm{i}a_1 h\varepsilon\Delta-\frac{1}{2}\boldsymbol{s}(t,h)^\top\nabla} = \mathrm{e}^{\mathrm{i}h\varepsilon a_1\partial_x^2-\frac{1}{2}s_x\partial_x}\mathrm{e}^{\mathrm{i}h\varepsilon a_1\partial_y^2-\frac{1}{2}s_y\partial_y},$$

where $\boldsymbol{s}=(s_x,s_y)$, can be implemented as

$$\mathcal{F}_x^{-1}\mathcal{D}_{\exp(\mathrm{i}h\varepsilon a_1 c_{2,x}-\frac{1}{2}s_x c_{1,x})}\mathcal{F}_x\mathcal{F}_y^{-1}\mathcal{D}_{\exp(\mathrm{i}h\varepsilon a_1 c_{2,x}-\frac{1}{2}s_y c_{1,x})}\mathcal{F}_y,$$

using the same number of FFTs[3].

In this way, the proposed method for time-dependent potentials of the form $V_0 + \boldsymbol{e}(t)^\top\boldsymbol{x}$ carries no additional expense compared to the order-four splitting of (Blanes & Moan 2002) for V_0 alone.

Remark 1 *Note that another variant of the splitting can be obtained by swapping the choices of A and B. In this variant the outermost exponential of (3.2) is not a Laplacian and, therefore, cannot be combined in this way with the outermost exponential of (3.1). In this case the proposed method requires one additional exponential.*

Remark 2 *An alternative, of course, is to perform any order-four splitting on the Magnus expansion $\Theta_2(t+h,t)$ directly by choosing $A=\mathrm{i}h\varepsilon\Delta-\frac{1}{2}\boldsymbol{s}^\top\nabla$ and $B=-\mathrm{i}h\varepsilon^{-1}\widetilde{V}(\boldsymbol{x},t,h)$. In light of the comments in this section, such variants also do not require additional expense in the case of discretisation via spectral collocation, for instance.*

4 A compact splitting of the Magnus expansion

As we have noted previously in subsection 3.1.2, the highly efficient compact splitting scheme of (Chin & Chen 2002),

$$\mathrm{e}^{X+Y} = \mathrm{e}^{\frac{1}{6}Y}\mathrm{e}^{\frac{1}{2}X}\mathrm{e}^{\frac{2}{3}(Y+\frac{1}{48}[[X,Y],Y])}\mathrm{e}^{\frac{1}{2}X}\mathrm{e}^{\frac{1}{6}Y} + \mathcal{O}\left(h^5\right). \tag{3.3}$$

for $X,Y=\mathcal{O}(h)$, features a central exponent which, in principle, has a nested commutator of X and Y. In practice, however, when X is the Laplacian term, this nested commutator reduces to a function, as we have seen in subsection 3.1.2. This results in a central exponent which is an $\mathcal{O}\left(h^3\right)$ perturbation of the potential function.

Since the relation (3.3) holds for arbitrary X,Y, in particular it also holds if we use it for splitting $\Theta_2(t+h,t)$ into the two parts

$$X=\mathrm{i}h\varepsilon\Delta-\boldsymbol{s}(t,h)^\top\nabla, \quad \text{and} \quad Y=-\mathrm{i}h\varepsilon^{-1}\widetilde{V}(\boldsymbol{x},t,h).$$

[3]Here $c_{1,x}, c_1 1, y, c_{2,x}$ and $c_{2,y}$ are the symbols of the differentiation matrices corresponding to $\partial_x, \partial_y, \partial_x^2$ and ∂_y^2, respectively.

Crucially, since the commutator of $\boldsymbol{s}^\top\nabla$ with a function $f(x)$ reduces to another function[4],

$$[\boldsymbol{s}^\top\nabla, f] = \sum_{j=1}^{n} s_j[\partial_{x_j}, f] = \sum_{j=1}^{n} s_j\left(f\partial_{x_j} + (\partial_{x_j}f) - f\partial_{x_j}\right) = \boldsymbol{s}^\top(\nabla f),$$

the nested commutator

$$[[\boldsymbol{s}^\top\nabla, f], f] = [\boldsymbol{s}^\top(\nabla f), f] = 0,$$

vanishes. The central exponent in this splitting is, therefore, identical to the one encountered in subsection 3.1.2,

$$\tfrac{2}{3}(Y + \tfrac{1}{48}[[X,Y],Y]) = -\tfrac{2}{3}\mathrm{i}h\varepsilon^{-1}\widehat{V} = -\tfrac{2}{3}\mathrm{i}h\varepsilon^{-1}(\widetilde{V} - \tfrac{h^2}{24}(\nabla\widetilde{V})^\top(\nabla\widetilde{V})).$$

Thus, a fourth-order compact splitting,

$$\mathrm{e}^{-\frac{1}{6}\mathrm{i}h\varepsilon^{-1}\widetilde{V}}\mathrm{e}^{\frac{1}{2}\mathrm{i}h\varepsilon\Delta-\frac{1}{2}\boldsymbol{s}(t,h)^\top\nabla}\mathrm{e}^{-\frac{2}{3}\mathrm{i}h\varepsilon^{-1}\widehat{V}}\mathrm{e}^{\frac{1}{2}\mathrm{i}h\varepsilon\Delta-\frac{1}{2}\boldsymbol{s}(t,h)^\top\nabla}\mathrm{e}^{-\frac{1}{6}\mathrm{i}h\varepsilon^{-1}\widetilde{V}}, \tag{4.1}$$

can be directly implemented using five stages (exponentials) for the case of time-dependent potentials.

This differs from the (Chin & Chen 2002) for time-independent potentials only in that a modified potential $\widetilde{V}$ is used in place of the time-independent potential V_0 and a perturbation of the Laplacian term, $\mathrm{i}h\varepsilon\Delta - \boldsymbol{s}(t,h)^\top\nabla$, is used instead of the usual $\mathrm{i}h\varepsilon\Delta$ occurring in (Chin & Chen 2002). In light of the remarks made in Subsection 3.1.3, neither of these add substantially to the cost of the method.

5 Numerical examples

In this section we will consider two one-dimensional numerical examples – the first in atomic scaling, $\varepsilon_1 = 1$, and the second in the semiclassical regime of $\varepsilon_2 = 10^{-2}$.

The initial conditions $u_{0,1}$ and $u_{0,2}$ for our numerical experiments are Gaussian wavepackets (with zero initial momentum),

$$u_{0,k}(x) = (\delta_k\pi)^{-1/4}\exp\left((-(x-x_0)^2)/(2\delta_k)\right), \quad x_0 = -2.5,\ k = 1,2,$$

with $\delta_1 = 0.2$ and $\delta_2 = 10^{-2}$ in the respective cases. These wavepackets are sitting in the left well of the double well potentials,

$$V_{\mathrm{D}1}(x) = x^4 - 15x^2 \quad \text{and} \quad V_{\mathrm{D}2}(x) = \tfrac{1}{5}x^4 - 2x^2,$$

respectively, which act as the choice of V_0 in the two examples.

Our spatial domain is $[-10,10]$ and $[-5,5]$ in the two examples, respectively, while the temporal domain is $[0,4]$ and $[0,\frac{5}{2}]$, respectively. We impose periodic boundaries on the spatial domains and resort to spectral collocation for discretisation.

When we allow the wave functions $u_{0,k}$ to evolve to $u_{\mathrm{D}k}$ at the final time, T, under the influence of $V_{\mathrm{D}k}$ alone, they remain largely confined to the left well (Figure 5.1, left column).

[4] By noting $[\partial_x, f]u = \partial_x(fu) - f(\partial_x u) = f\partial_x u + (\partial_x f)u - f(\partial_x u) = (\partial_x f)u$, we conclude the operatorial identity $[\partial_x, f] = (\partial_x f)$.

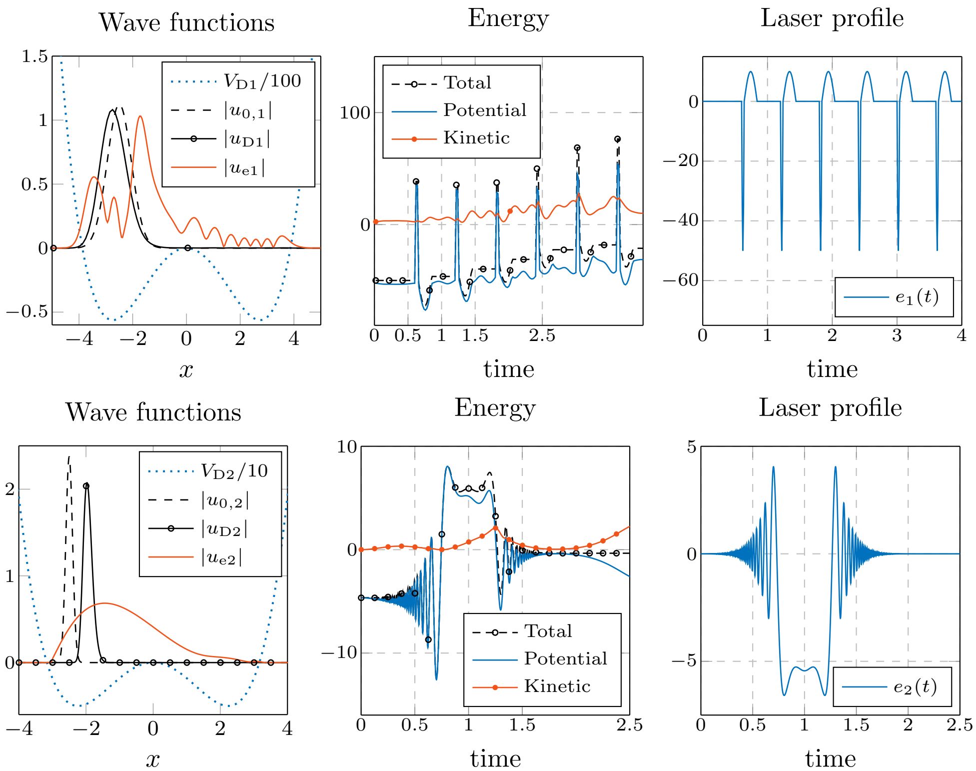

Figure 5.1: [Example 1 (top row), Example 2 (bottom row)]. The initial condition $u_{0,k}$ evolves to $u_{\mathrm{D}k}$ at time T under the influence of $V_0 = V_{\mathrm{D}k}$ and to $u_{\mathrm{e}k}$ under $V_{\mathrm{e}k} = V_{\mathrm{D}k} + e_k(t)x$ (left); Evolution of energy under $V_{\mathrm{e}k}$ (centre); Laser profile $e_k(t)$ (right). Here, the potentials $V_{\mathrm{D}k}$ are scaled down for ease of presentation, $k = 1$ for Example 1 and $k = 2$ for Example 2.

Time-dependent potential. Superimposing a time dependent excitation of the form $e(t)\,x$ on the potential – used for modeling laser interaction – we are able to exert control on the wave function. The time profile of the laser used here is

$$e_1(t) = \begin{cases} \sin(25\pi t) & t \in [\frac{3}{5}n, \frac{3}{5}n + \frac{1}{25}], \quad n \geq 1, \\ \sin(5\pi t) & t \in (\frac{3}{5}n + \frac{1}{25}, \frac{3}{5}n + \frac{6}{25}], \quad n \geq 1, \end{cases}$$

and

$$e_2(t) = 10\exp(-10(t-1)^2)\sin((500(t-1)^4 + 10)),$$

respectively. The former is a sequence of *asymmetric sine lobes* while the latter is a highly oscillatory *chirped* pulse (Figure 5.1, right column). Such laser profiles are used routinely in laser control (Amstrup et al. 1993). Even more oscillatory electric fields often result from optimal control algorithms (Meyer et al. 2006, Coudert 2018).

The effective time-dependent potentials in the two examples are

$$V_{\mathrm{e}k}(x,t) = V_{\mathrm{D}k}(x) + e_k(t)x, \quad k = 1, 2.$$

Under the influence of this time-dependent potential $V_{\mathrm{e}k}(x,t)$, the initial wave-functions $u_{0,k}$ evolve to $u_{\mathrm{e}k}$, which are not confined to the left well (Figure 5.1, left column). In this case the total energy is not conserved (Figure 5.1, middle column).

Methods. We denote the combination of the fourth-order splitting (3.2) with (3.1), which is derived in Subsection 3.1.1, by MaStBM (Magnus–Strang–Blanes–Moan). This method implements the outermost exponential in (3.1) separately and is expected to be the most costly of our schemes. As noted in Subsection 3.1.3, the outermost exponentials can be combined, resulting in a method which will be labeled as MaStBMc (c for combined).

Another concrete example of our splittings is outlined in Subsection 3.1.2, where a more efficient method (3.6) results by resorting to (Chin & Chen 2002) for the approximation of the central exponent in (3.1). This scheme is denoted by MaStCC (Magnus–Strang–Chin–Chen). It requires computation of an additional exponential in comparison with the method of (Chin & Chen 2002). The most efficient method in this class (4.1), results from the direct application of (Chin & Chen 2002) to Θ_2 in Section 4, and is denoted by MaCC (Magnus–Chin–Chen). This method should cost no more than an implementation of the method from (Chin & Chen 2002), which, of course, is designed for time-independent potentials. In this way, the fourth order schemes presented here are able to handle time-dependent potentials with little or no additional costs.

To demonstrate the effectiveness of our fourth-order schemes, we will compare them to a couple of sixth-order methods. The first of these is the sixth-order optimised method CF6:5Opt proposed in (Alvermann & Fehske 2011), which is labeled as AF in this section. AF is accompanied by a postfix which refers to the number of Lanczos iterations (5, 10 or 50) used in the method. The second scheme is the sixth-order method of (Iserles et al. 2018*b*), denoted by IKS, which is specialised for the semiclassical regime. In all cases presented here IKS, utilises three Lanczos iterations for the exponentiation of its first non-trivial exponent, $W^{[2]}$, and two Lanczos iterations for the exponentiation of the innermost exponent, $\mathcal{W}^{[3]}$.

The first of these methods, AF, is not specialised for laser potentials, highly oscillatory potentials or for the semiclassical regime. The second, IKS, is optimised for highly oscillatory potentials and does provide certain optimisations for the case of lasers, which have been employed in these experiments. However, it is not designed for atomic scaling.

In the first example, the integrals in our schemes, (2.4) and (2.5), as well as those in IKS are discretised via three Gauss–Legendre knots, while in the second case eleven Gauss–Legendre knots are used in order to adequately resolve the highly oscillatory potential. Not discretising these integrals at the outset allows us flexibility in deciding a quadrature strategy at the very end, as discussed by (Iserles et al. 2018*a*, Iserles et al. 2018*b*). Effectively, this strategy allows us to resolve a highly oscillatory potential despite using large time steps in the propagation of the solution. In contrast, the method AF uses a fixed number (four) of Gauss–Legendre knots for all cases.

In the first example $\varepsilon_1 = 1$, we use $M_1 = 150$ spatial grid points while the highly

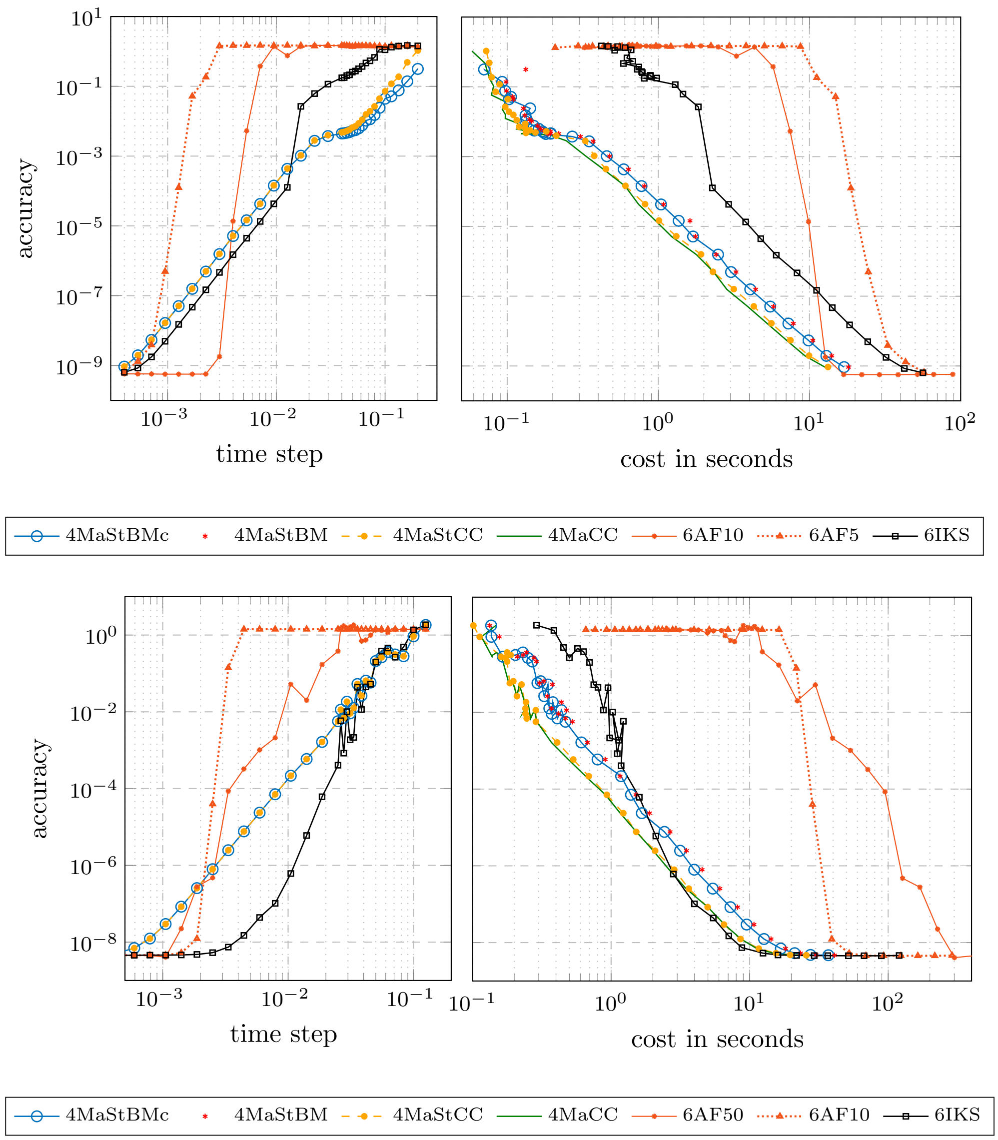

Figure 5.2: [Example 1 (top row), Example 2 (bottom row)] Accuracy *vs* time step (left); accuracy *vs* cost in seconds (right). The prefixes 4 and 6 are used to highlight that these are order 4 and order 6 methods, respectively.

oscillatory behaviour in the second example, that occurs because of the small semi-classical parameter $\varepsilon_2 = 10^{-2}$, requires $M_2 = 2000$ spatial grid points.

Remark 3 *Even though IKS behaves like an order four method for* $\varepsilon = 1$*, it is surpris-*

ing that it works as well as it does considering that it is designed for the semiclassical regime $\varepsilon \ll 1$. In particular it outperforms the sixth-order AF for large time steps.

Reference solutions. The reference solutions for these experiments were generated by using the sixth-order scheme AF10 with very small time steps. In both examples we use $M = 5000$ grid points and $h = T/10^6$ as time step, which corresponds to 10^6 time steps. As usual, the L^2 distance from the reference solution is used as a measure of accuracy in these experiments.

6 Conclusions

To summarise, in Sections 2 and 3, we have presented a general strategy for quickly extending any existing fourth-order method for time-independent potentials to effectively handle the case of laser potentials. The overall schemes require, at most, one additional exponential, which leads to a very marginal increase in cost.

Further optimisation steps are possible which allow us to use exactly the same number of exponentials as the fourth-order methods for time-independent potentials. In particular, in Sections 2 and 4, we have presented a highly optimised compact splitting that requires merely five exponentials, requiring 4 FFTs (per dimension) per step.

As we can see from the numerical results presented in Figure 5.2, these fourth-order schemes usually exceed the accuracy of the sixth-order methods of (Alvermann & Fehske 2011) for moderate to large time steps, while being significantly cheaper. Although the specialised sixth-order approach of (Iserles et al. 2018*b*) has a very high accuracy in the semiclassical regime (as one might expect), the low costs of our methods makes them highly competitive with that scheme. The ease of implementing the proposed schemes or extending existing implementations should make this strategy appealing.

Note that, these methods are not derived under any specific spatial discretisation choice. The assumption of spectral collocation in Subsection 3.1.3 serves only to concretely demonstrate the fact that certain exponentials can be combined in practice. In principle, it should be possible to utilise these schemes for strategies such as Hagedorn wavepackets (Gradinaru & Hagedorn 2014), which allow computation over the real line, or the use of absorbing boundaries, which might be helpful in the case of non-confining potentials.

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