Magnus–Lanczos methods with simplified commutators for the Schrödinger equation with a time-dependent potential

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Abstract

The computation of the Schrödinger equation featuring time-dependent potentials is of great importance in quantum control of atomic and molecular processes. These applications often involve highly oscillatory potentials and require inexpensive but accurate solutions over large spatio-temporal windows. In this work we develop Magnus expansions where commutators have been simplified. Consequently, the exponentiation of these Magnus expansions via Lanczos iterations is significantly cheaper than that for traditional Magnus expansions. At the same time, and unlike most competing methods, we simplify integrals instead of discretising them via quadrature at the outset – this gives us the flexibility to handle a variety of potentials, being particularly effective in the case of highly oscillatory potentials, where this strategy allows us to consider significantly larger time steps.

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1 Introduction

We consider the linear, time-dependent Schrödinger equation (TDSE) for a single particle moving in a time-varying electric field,

$$\frac{\partial u(x,t)}{\partial t} = \mathrm{i}\frac{\partial^2 u(x,t)}{\partial x^2} - \mathrm{i}V(x,t)u(x,t), \quad x \in \mathbb{R},\ t \geq 0, \tag{1.1}$$

where the complex-valued wave function $u = u(x,t)$ is given with an initial condition $u(x,0) = u_0(x)$. Here $V(x,t)$ is a real-valued, time-dependent electric field, and we are working in atomic units, where Planck's constant is scaled to one ($\hbar = 1$).

These equations are of great practical importance since they allow us to study the behaviour of particles under the influence of changing electrical field. As our ability to manipulate electric fields becomes more refined, including the shaping of laser pulses, unprecedented quantum control of atomic and molecular systems is becoming possible (Shapiro & Brumer 2003). Optimal control of quantum systems is among the many challenges that require highly accurate and computationally inexpensive solutions of this equation, often involving highly oscillatory potentials over large spatio-temporal windows

1.1 Existing approaches

Time-dependent potentials significantly complicate matters insofar as numerical solutions are concerned. Typically the solution of (1.1) involves a truncation of the Magnus expansion, which is an infinite series of nested integrals of nested commutators, as we will see in this section.

Traditional methods for solving (1.1) usually commence with spatial discretisation,

$$\boldsymbol{u}'(t) = \mathrm{i}(\mathcal{K}_2 - \mathcal{D}_{V(\cdot,t)})\boldsymbol{u}(t), \qquad t \geq 0, \tag{1.2}$$

where the vector $\boldsymbol{u}(t) \in \mathbb{C}^M$ represents an approximation to the solution at time t, $\boldsymbol{u}(0) = \boldsymbol{u}_0$ is derived from the initial conditions, while $\mathcal{K}_2$ and $\mathcal{D}_{V(\cdot,t)}$ are $M \times M$ matrices which represent (discretisation of) second derivative and a multiplication by the interaction potential $V(\cdot,t)$, respectively.

Magnus expansions. The system of ODEs (1.2), which is of the form

$$\boldsymbol{u}'(t) = A(t)\boldsymbol{u}(t), \qquad t \geq 0, \tag{1.3}$$

with $A(t) = \mathrm{i}(\mathcal{K}_2 - \mathcal{D}_{V(\cdot,t)})$, can be solved via the Magnus expansion (Magnus 1954),

$$\boldsymbol{u}(t) = \mathrm{e}^{\Theta(t,s)}\boldsymbol{u}(s), \tag{1.4}$$

where $\Theta(t,s)$ is a time-dependent $M \times M$ skew-Hermitian matrix whose exponential evolves the solution from time s to t. The Magnus expansion $\Theta(t,s)$ is obtained as an infinite series $\sum_{k=1}^{\infty} \Theta^{[k]}(t,s)$ with each $\Theta^{[k]}(t,s)$ composed of k nested integrals and $k-1$ nested commutators (see expressions below).

In practice, we work with finite truncations of the Magnus series,

$$\Theta_m(t,s) = \sum_{k=1}^{m} \Theta^{[k]}(t,s),$$

and propagate the solution in suitably small time steps h,

$$\boldsymbol{u}^{n+1} = \mathrm{e}^{\Theta_m(t_n+h,t_n)}\boldsymbol{u}^n,$$

in order to keep the truncation error low. For the sake of simplicity, we analyse only the first step,

$$\boldsymbol{u}^1 = \mathrm{e}^{\Theta_m(h)}\boldsymbol{u}^0, \tag{1.5}$$

writing $\Theta_m(h) = \Theta_m(h,0)$, for short[1]. The first few terms of $\Theta_m(h)$ are

$$\begin{aligned}
\Theta^{[1]}(h) &= \int_0^h A(\xi_1)d\xi_1,\\
\Theta^{[2]}(h) &= -\frac{1}{2}\int_0^h \left[\int_0^{\xi_1} A(\xi_2)d\xi_2, A(\xi_1)\right] d\xi_1,\\
\Theta^{[3]}(h) &= \frac{1}{12}\int_0^h \left[\int_0^{\xi_1} A(\xi_2)d\xi_2, \left[\int_0^{\xi_1} A(\xi_2)d\xi_2, A(\xi_1)\right]\right] d\xi_1\\
&\quad + \frac{1}{4}\int_0^h \left[\int_0^{\xi_1} \left[\int_0^{\xi_2} A(\xi_3)d\xi_3, A(\xi_2)\right] d\xi_2, A(\xi_1)\right] d\xi_1.
\end{aligned}$$

Exponential midpoint rule. The simplest method in this family results from letting $\Theta_1(h) = \Theta^{[1]}(h)$,

$$\boldsymbol{u}^1 = \mathrm{e}^{\Theta_1(h)}\boldsymbol{u}^0 = \exp\left(\int_0^h A(\xi)d\xi\right)\boldsymbol{u}^0 = \exp\left(\mathrm{i}h\mathcal{K}_2 - \mathrm{i}\int_0^h \mathcal{D}_{V(\xi)}d\xi\right)\boldsymbol{u}^0.$$

This method, called the exponential midpoint rule, is well known and has been used for a long while (Tal-Ezer, Kosloff & Cerjan 1992, Lubich 2008). It is typical to approximate $\int_0^h \mathcal{D}_{V(\xi)}d\xi = \mathcal{D}_{\int_0^h V(\xi)d\xi}$ by taking the value of V at the middle of the integral, $\int_0^h V(\xi)d\xi \approx hV(h/2)$, and concluding with an application of the Strang splitting,

$$\boldsymbol{u}^1 = \exp\left(\tfrac{1}{2}\mathrm{i}h\mathcal{K}_2\right)\exp\left(-\mathrm{i}h\mathcal{D}_{V(h/2)}\right)\exp\left(\tfrac{1}{2}\mathrm{i}h\mathcal{K}_2\right)\boldsymbol{u}^0. \tag{1.6}$$

Remark 1 *The* power-truncated *Magnus expansions used here are time-symmetric and can be expanded solely in odd powers of h (Iserles & Nørsett 1999, Iserles, Munthe-Kaas, Nørsett & Zanna 2000).*

Remark 2 *We note that the first truncation, Θ_1, carries an error of $\mathcal{O}\left(h^3\right)$ (Iserles et al. 2000). Ideally it should be combined with an $\mathcal{O}\left(h^3\right)$ quadrature. However, due to Remark 1, an $\mathcal{O}\left(h^{2n}\right)$ accuracy quadrature method will have an accuracy of $\mathcal{O}\left(h^{2n+1}\right)$ in this context, resulting in the need for fewer quadrature points.*

[1]The corresponding solution for any step $\mathrm{e}^{\Theta_m(t_n+h,t_n)}\boldsymbol{u}^n$ can be obtained by replacing $A(\zeta)$ by $A(t_n+\zeta)$ in the Magnus expansion.

As a consequence of Remark 2 and the $\mathcal{O}\left(h^3\right)$ accuracy of the Strang splitting, (1.6) has a local error of $\mathcal{O}\left(h^3\right)$.

Higher order truncations of the Magnus expansion. Once higher order accuracy is desired (Tal-Ezer et al. 1992, Kormann, Holmgren & Karlsson 2008), we need to consider higher order truncations of the Magnus expansion such as

$$\Theta_2(h) = \int_0^h A(\xi_1)d\xi_1 - \frac{1}{2}\int_0^h \left[\int_0^{\xi_1} A(\xi_2)d\xi_2, A(\xi_1)\right] d\xi_1 = \Theta(h) + \mathcal{O}\left(h^5\right).$$

Higher order truncations necessarily involve nested integrals of nested commutators. The nested integrals here need to be approximated using quadrature formulæ of accuracy $\mathcal{O}\left(h^5\right)$. However, as mentioned before, it suffices to consider the Gauss–Legendre quadrature at only two nodes: $\tau_k = \frac{h}{2}(1 \pm 1/\sqrt{3})$. This results in the method

$$\boldsymbol{u}^1 = \exp\Big(\tfrac{h}{2}(A(\tau_{-1}) + A(\tau_1)) + \tfrac{\sqrt{3}h^2}{12}[A(\tau_{-1}), A(\tau_1)]\Big)\,\boldsymbol{u}^0.$$

For the Schrödinger equation (1.1), this translates to

$$\boldsymbol{u}^1 = \exp\Big(\mathrm{i}h\mathcal{K}_2 - \mathrm{i}h\overline{V} + h^2\tfrac{\sqrt{3}h^2}{12}[\mathcal{K}_2, \widetilde{V}]\Big)\,\boldsymbol{u}^0.$$

where $\overline{V} = [V(\tau_{-1}) + V(\tau_1)]/2$ and $\widetilde{V} = V(\tau_{-1}) - V(\tau_1)$.

Splitting the exponential of Magnus expansions. The exponential of Θ_2 needs to be evaluated up to an accuracy of $\mathcal{O}\left(h^5\right)$. The second-order Strang splitting,

$$\mathrm{e}^{\Theta_2(h)} = \mathrm{e}^{\frac{1}{2}\mathrm{i}h\mathcal{K}_2}\ \mathrm{e}^{-\frac{1}{2}\mathrm{i}h\mathcal{D}_{\overline{V}}}\ \mathrm{e}^{h^2\frac{\sqrt{3}h^2}{12}[\mathcal{K}_2,\mathcal{D}_{\widetilde{V}}]}\ \mathrm{e}^{-\frac{1}{2}\mathrm{i}h\mathcal{D}_{\overline{V}}}\ \mathrm{e}^{\frac{1}{2}\mathrm{i}h\mathcal{K}_2} + \mathcal{O}\left(h^3\right),$$

therefore, does not suffice. Instead we require the fourth order Yoshida splitting, obtained by composing three order-two Strang splittings.

When the exponent to be split consists of two components, the number of exponentials in an order-$2m$ Yoshida splitting grows as $2 \times 3^{m-1} + 1$. Here, we need to approximate the exponential of higher order Magnus truncations, Θ_m, which feature an increasingly larger number of terms. Consequently the number of exponentials in the Yoshida splitting for Magnus expansions grows even more rapidly.

Moreoever, we are left with the problem of evaluating the exponential of commutators such as $[\mathcal{K}_2, \mathcal{D}_{\widetilde{V}}]$ which are expensive to compute and do not posses a structure that allows for an easy exponentiation. In higher-order methods such as Θ_3 we start encountering commutators in a nested form.

Magnus–Lanczos schemes. An alternative approach for approximating the exponential of the Magnus expansion is via Lanczos iterations (Gallopoulos & Saad 1992), leading to the popular Magnus–Lanczos schemes. This is, arguably, a more flexible approach since we only require a method for computing matrix–vector products of the form $\Theta_n v$ in each Lanczos iteration.

Nevertheless the exponential growth resulting from the presence of nested commutators is inevitable. Moreover, the highly promising superlinear decay of error in the case of the Lanczos method for approximating the matrix exponential is not seen until the number of iterations is larger than the spectral radius of $\Theta_m(h)$ (Hochbruck & Lubich 1997), which is very large unless the time step h is suitably small.

Commutator-free, integral-free quasi-Magnus methods. To avoid the exponential growth of cost due to presence of nested commutators, many attempts have been made at deriving commutator-free schemes. These usually proceed by replacing nested integrals in the Magnus expansion by some quadratures or Taylor expansions of V at the outset, subsequently seeking a commutator-free exponential splitting that adequately approximates the exponential of the discretised Magnus expansion. Since the Magnus expansion does not appear explicitly in these schemes, they are sometimes also referred to as *quasi-Magnus*.

For example in (Alvermann & Fehske 2011), instead of the exponential of Magnus expansion, authors derive an alternative numerical propagator for Schrödinger equations, namely a product of exponentials of linear combinations of various values of Hamiltonian operator (more precisely, values of Hamiltonian operator are taken in Gauss-Legendre quadrature points). Blanes, Casas & Thalhammer (2017*b*), on the other hand, investigate the commutator-free expansion for differential equations of both parabolic and hyperbolic equations, also providing stability and error analysis.

Integral-free Magnus–Zassenhaus splittings with simplified commutators. In (Bader, Iserles, Kropielnicka & Singh 2016) an integral-free numerical integrator with simplified commutators was proposed for Schrödinger equation in the semiclassical regime. Once again, these proceed via discretisation of the integrals in the Magnus expansion. However, unlike the commutator-free methods where commutators are eliminated, these work by simplifying the commutators in the Lie algebra of anti-commutators, subsequently exploting the idea of the symmetric Zassenhaus asymptotic splittings (Bader, Iserles, Kropielnicka & Singh 2014) for exponentiation. These have been shown to be highly effective in the semiclassical regime.

Other notable approaches. Expansion in Chebyshev polynomials is an effective alternative to Lanczos iterations for approximating the exponential of the Magnus expansion, particularly when large time steps are involved (Tal-Ezer et al. 1992, Ndong, Tal-Ezer, Kosloff & Koch 2010, Tal-Ezer, Kosloff & Schaefer 2012, Schaefer, Tal-Ezer & Kosloff 2017). Other competing approaches that forego Magnus expansions entirely include polynomial approximations to the propagator based on Taylor expansions (Lauvergnat, Blasco, Chapuisat & Nauts 2007), symplectic splitting methods (Blanes, Casas & Murua 2017*a*), Runge–Kutta methods (Tremblay & Carrington Jr. 2004), symplectic partitioned Runge–Kutta methods (Sanz-Serna & Portillo 1996) and the (t, t') method (Peskin, Kosloff & Moiseyev 1994).

1.2 Main contributions

In this work we present *Magnus–Lanczos methods with simplified commutators* that

1. retain all the advantages of Magnus expansions and Lanczos methods,
2. are free of nested commutators (and the associated growth in cost),
3. feature non-nested anti-commutators which preserve skew-Hermiticity of the expansion (thus unitarity of solution and stability of the discretised method),
4. preserve the integrals intact until the very last moment of the algorithm (this enables more flexibility, higher accuracy and often lower cost while dealing with numerical integration),

5. feature fewer nested integrals (due to identities (2.11) and (2.12)). Our order six methods, for instance, feature only twice-nested integrals instead of the quadruply-nested integrals that feature in a standard Magnus expansion – not only is this beneficial for numerical quadrature, but it also makes analytic and asymptotic approximation easier.

As discussed previously, standard Magnus expansions feature nested commutators. When we need to compute $\Theta_m v$ in each Lanczos iteration, these nested commutators result in the cost of $\Theta_m v$ growing exponentially in m. The absence of nested commutators in our Magnus expansions, where such commutators have been simplified, results in the cost of $\Theta_m v$ growing linearly in m.

Moreover, we are able to do this while keeping integrals intact, resulting in methods that are highly flexible – not only it is possible to approximate the integrals through any quadrature method, but we may also use exact integrals for potentials whenever possible. This proves particularly effective in the case of potentials with high temporal oscillations where we no longer require a severe depression of time steps.

1.3 Organisation of the paper

Section 2 is devoted to the simplification of commutators in the Magnus expansion. Magnus expansions for the Schrödinger equation evolve in the Lie algebra generated by ∂_x^2 and $V(\cdot)$. However, as it will be pointed out in Subsection 2.1, a straightforward simplification of commutators of these operators using the chain rule results in the loss of unitarity of the solution upon discretisation. One of the novelties of our approach is working in the algebra of anti-commutators, which leads to the preservation of skew-Hermitian structure and stability of the scheme. The procedure for deriving Magnus expansions with simplified commutators is presented in Subsections 2.2–2.6. In Subsections 2.7 and 2.8 we present concrete order four and order six Magnus expansions, Θ_2 and Θ_4, respectively (methods (2.19) and (2.26)).

Section 3 deals with the implementation of our method. In Subsection 3.1 we provide some details concerning spatial discretisation. Subsection 3.2 deals with the evaluation of derivatives and integrals of the potential appearing in the Magnus expansion. While various alternatives are possible at this stage, a particular option – namely, finite differences for derivatives and Gauss–Legendre quadrature with three knots for integrals – is outlined in greater detail (expressions 3.3–3.8). In Subsection 3.3 we discuss the implementation of Lanczos iterations (achieved via (3.9)) for numerical exponentiation of the Magnus expansion and the number of Fast Fourier Transforms (FFTs) required per iteration.

Numerical examples are provided in Section 4, while in the last section we briefly summarise our results.

2 Magnus expansions with simplified commutators

In contrast to the traditional approach of resorting to spatial discretisation of (1.1), which leads to the system of finite dimensional ODEs (1.2) followed by the Magnus expansion (1.4), we begin straight away with a Magnus expansion of (1.1) while keeping the underlying operators intact.

In numerically solving (1.1), consistently with standard practice we impose periodic boundary conditions on a finite interval $I \subseteq \mathbb{R}$. We further assume throughout that the interaction potential $V(\cdot, t)$ and the wavefunction $u(\cdot, t)$ are sufficiently smooth. For the purpose of this paper and for simplicity sake we assume that they belong to $\mathrm{C}_p^\infty(I;\mathbb{R})$ and $\mathrm{C}_p^\infty(I;\mathbb{C})$, respectively, the spaces of real valued and complex valued smooth periodic functions over I, but our results extend in a straightforward manner to functions of lower smoothness (the regularity constraints will depend on the desired order of the method being derived).

Considering (1.1) as an evolutionary PDE evolving in a Hilbert space, say $\mathcal{H} = \mathrm{L}^2(I;\mathbb{C})$, and suppressing the dependence on x,

$$\partial_t u(t) = \left(\mathrm{i}\partial_x^2 - \mathrm{i}V(t)\right) u(t), \quad u(0) = u_0 \in \mathcal{H}, \tag{2.1}$$

is seen to be of the 'ODE-like' form

$$\partial_t u(t) = A(t)u(t), \quad u(0) = u_0 \in \mathcal{H}, \tag{2.2}$$

with $A(t) = \mathrm{i}\partial_x^2 - \mathrm{i}V(t)$. The operator $A(t)$ belongs to $\mathfrak{u}(\mathcal{H})$, the Lie algebra of (infinite-dimensional) skew-Hermitian operators acting on the Hilbert space $\mathcal{H}$. Its flow is, therefore, unitary and resides in $\mathcal{U}(\mathcal{H})$ – the Lie group of unitary operators.

Unitary evolution of the wave function $u(t)$ under this flow is fundamental to quantum mechanics. Preservation of this property under discretisation is very important and we seek appropriate geometric numerical integrators to guarantee it. This comes about naturally once we work in the correct Lie-algebraic framework. As we note later, unitarity also guarantees stability of a consistent numerical scheme.

For a general equation of the form (2.2) where $A(t)$ resides in a Lie algebra $\mathfrak{g}$, the solution for the flow can be formally written in the form of a Magnus expansion,

$$u(h) = \mathrm{e}^{\Theta(h)} u(0), \tag{2.3}$$

which differs from (1.4) in the sense that the Magnus expansion Θ is in general an infinite-dimensional and unbounded operator, not a matrix.

Remark 3 *Convergence of the Magnus expansion, in the finite dimensional case is only guaranteed for sufficiently small time steps (Moan & Niesen 2008). In principle, this becomes problematic when we consider Magnus expansions of undiscretised and unbounded operators. An extension to bounded operators on an infinite-dimensional Hilbert space was done by (Casas 2007).*

Rigorous analysis in the context of the Schrödinger equation, which features an unbounded operator, has been carried out by (Hochbruck & Lubich 2002) who show that when Magnus expansions of unbounded operators are considered in a formal sense, the concrete methods based on these approaches do demonstrate the expected order of convergence.

Pursuing a similar strategy, it is possible to derive rigorous error bounds for the Magnus expansion based methods presented in this manuscript. However, since this analysis involves development of additional theory that could obscure the presentation of the proposed methods, it will be beyond the scope of our investigations. Here we refer the curious reader to Chapter 9. of (Singh 2017).

2.1 The algebra of anti-commutators

The vector field in the Schrödinger equation (2.1) is a linear combination of the action of two operators, ∂_x^2 and the multiplication by the interaction potential $V(t)$, for any $t \geq 0$. Since the Magnus expansion requires nested commutation, the focus of our interest is the Lie algebra generated by ∂_x^2 and $V(\cdot)$,

$$\mathfrak{F} = \mathrm{LA}\{\partial_x^2, V(\cdot)\},$$

i.e. the linear-space closure of all nested commutators of ∂_x^2 and $V(\cdot)$.

Simplifying commutators. To simplify commutators, we follow the approach of (Bader et al. 2014) and study their action on functions. For example, using the chain rule we find,

$$[\partial_x^2, V]u = \partial_x^2(Vu) - V(\partial_x^2 u) = (\partial_x^2 V)u + 2(\partial_x V)\partial_x u,$$

which implies that $[\partial_x^2, V] = (\partial_x^2 V) + 2(\partial_x V)\partial_x$. Similarly, we conclude that

$$\begin{aligned} [\partial_x^2, [\partial_x^2, V]] &= (\partial_x^4 V) + 4(\partial_x^3 V)\partial_x + 4(\partial_x^2 V)\partial_x^2, \\ [V, [\partial_x^2, V]] &= -2(\partial_x V)^2. \end{aligned}$$

Note that we have ignored here the dependence on the time variable since the derivatives are only in the spatial variable.

Loss of skew-Hermiticity. Simplifying commutators in this way, we can, in principle, get rid of all nested commutators occurring in the (truncated) Magnus expansion of the undiscretised operators. It is only after this stage that we would resort to spatial discretisation. Proceeding in this way, however, we break an important structural property – upon discretisation, such a Magnus expansion is no longer skew-Hermitian and thus its exponential is no longer unitary.

To illustrate the loss of unitarity, let us consider the two differential operators: $[\partial_x^2, V]$ and $(\partial_x^2 V) + 2(\partial_x V)\partial_x$. Spatial discretisation transforms these two analytically identical operators to $[\mathcal{K}_2, \mathcal{D}_V]$ and $\mathcal{D}_{\partial_x^2 V} + 2\mathcal{D}_{\partial_x V}\mathcal{K}_1$, respectively. Assuming that $\mathcal{K}_n$ is skew-symmetric for odd n and symmetric for even n (i.e. the skew-symmetry of ∂_x is preserved under discretisation) and $\mathcal{D}_V$ is symmetric (note that V is real-valued and $\mathcal{D}_V$ represents multiplication by V), the commutator $[\mathcal{K}_2, \mathcal{D}_V]$ is skew-symmetric. However, the second expression, subject to discretisation, is no longer skew-symmetric. A similar problem is encountered in the discretisation of $[\partial_x^2, [\partial_x^2, V]]$ following the simplification of the commutator.

The loss of skew-symmetry in the simplification of $[\partial_x^2, V]$ (and skew-Hermiticity in general) is a cause for concern on two accounts: firstly, the exponential of a Magnus expansion which features terms like $[\partial_x^2, V]$ (Θ_2, for instance) is no longer unitary, which is highly undesirable insofar as the physics is concerned; secondly, since $\mathcal{D}_{\partial_x^2 V} + 2\mathcal{D}_{\partial_x V}\mathcal{K}_1$ has (large) real eigenvalues, its exponential blows up, which is highly undesirable from the numerical point of view. This blowup can be extreme even in the simplest of cases (see Figure 2.1).

Remark 4 *Note that* $[\partial_x^2, V]$ *and the simplified form* $(\partial_x^2 V) + 2(\partial_x V)\partial_x$ *are both skew-Hermitian operators. However, the straightforward discretisation (i.e. where every*

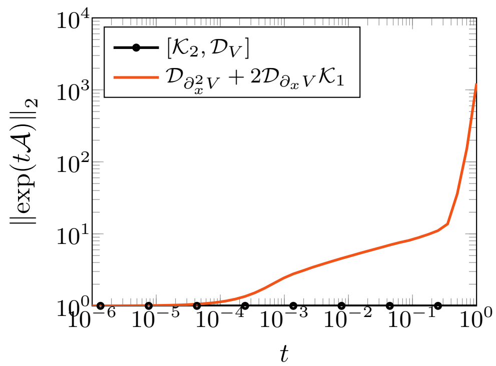

Figure 2.1: The two equivalent forms $[\partial_x^2, V]$ and $(\partial_x^2 V) + 2(\partial_x V)\partial_x$, lead to two different discretisations, $[\mathcal{K}_2, \mathcal{D}_V]$ and $\mathcal{D}_{\partial_x^2 V} + 2\mathcal{D}_{\partial_x V}\mathcal{K}_1$. While the exponential of the former is unitary, the exponential of the latter blows up.

instance of $V^{(k)}$ is replaced by $\mathcal{D}_{V^{(k)}}$ and ∂_x^k by $\mathcal{K}_k$) of the simplified form to $\mathcal{D}_{\partial_x^2 V} + 2\mathcal{D}_{\partial_x V}\mathcal{K}_1$ is where skew-Hermiticity is lost. This is not entirely surprising since a discretisation scheme, in general, might respect the skew-Hermiticity of only a subset of skew-Hermitian operators.

The Lie algebra of anti-commutators. In the methods presented here we circumvent the problem of skew-Hermitian discretisation by working with differential operators of the form

$$\langle f \rangle_k := \tfrac{1}{2}\left(f \circ \partial_x^k + \partial_x^k \circ f\right), \quad k \geq 0,\ f \in \mathrm{C}_p^\infty(I;\mathbb{R}), \tag{2.4}$$

which are inherently symmetrised. These are anti-commutators of f and ∂_x^k. The action of this differential operator on u, for example, is

$$\langle f \rangle_k u = \tfrac{1}{2}\left(f\partial_x^k u + \partial_x^k(fu)\right)$$

and the discretisation of this operator is

$$\langle f \rangle_k \rightsquigarrow \tfrac{1}{2}\left(\mathcal{D}_f \mathcal{K}_k + \mathcal{K}_k \mathcal{D}_f\right). \tag{2.5}$$

Remark 5 *It is a very simple process to verify that $\langle f \rangle_k$ is a skew-Hermitian operator for odd k and Hermitian operator for even k. This feature is maintained under straightforward discretisation. This is the reason why the choice of algebra of anti-commutators $\langle \cdot \rangle_k$ seems to be optimal for our purposes.*

The commutators of these operators can be solved using the following rules,

$$
\begin{aligned}
[\langle f\rangle_1, \langle g\rangle_0] &= \langle f(\partial_x g)\rangle_0, \\
[\langle f\rangle_1, \langle g\rangle_1] &= \langle f(\partial_x g) - (\partial_x f)g\rangle_1, \\
[\langle f\rangle_2, \langle g\rangle_0] &= 2\langle f(\partial_x g)\rangle_1, \\
[\langle f\rangle_2, \langle g\rangle_1] &= \langle 2f(\partial_x g) - (\partial_x f)g\rangle_2 - \tfrac{1}{2}\left\langle 2(\partial_x f)(\partial_x^2 g) + f(\partial_x^3 g)\right\rangle_0, \\
[\langle f\rangle_2, \langle g\rangle_2] &= 2\langle f(\partial_x g) - (\partial_x f)g\rangle_3 + \left\langle 2(\partial_x^2 f)(\partial_x g) - 2(\partial_x f)(\partial_x^2 g) + (\partial_x^3 f)g - f(\partial_x^3 g)\right\rangle_1, \\
[\langle f\rangle_3, \langle g\rangle_0] &= 3\langle f(\partial_x g)\rangle_2 - \tfrac{1}{2}\left\langle 3(\partial_x f)(\partial_x^2 g) + f(\partial_x^3 g)\right\rangle_0, \\
[\langle f\rangle_4, \langle g\rangle_0] &= 4\langle f(\partial_x g)\rangle_3 - 2\left\langle 3(\partial_x f)(\partial_x^2 g) + f(\partial_x^3 g)\right\rangle_1.
\end{aligned}
\tag{2.6}
$$

Remark 6 *Note that each commutator is simplified to a linear combination of $\langle\cdot\rangle_k$s where the indices k are either even or odd, but the two are never mixed. Upon discretisation these individual terms will result in either Hermitian matrices or skew-Hermitian matrices, but never a mixture of the two. Moreover, any term can be discarded (say, due to a small size) without disturbing the symmetry.*

There is rich algebraic theory underlying these operators (including a general formula for (2.6)) which feature in a separate publication (Singh 2015). In principle, however, the above rules can be verified by application of the chain rule.

Remark 7 *Note that, by definition (2.4),*

1. *these brackets are linear, so that $\langle 2f(\partial_x g) - (\partial_x f)g\rangle_2 = 2\langle f(\partial_x g)\rangle_2 - \langle(\partial_x f)g\rangle_2$,*
2. *$\langle f\rangle_0 = f$; and*
3. *$\langle 1\rangle_2 = \partial_x^2$.*

With this new notation in place and using (2.6), we can now simplify commutators to anti-commutators,

$$
\begin{aligned}
[\mathrm{i}\partial_x^2, \mathrm{i}V] &= -[\langle 1\rangle_2, \langle V\rangle_0] = -2\langle \partial_x V\rangle_1, \\
[\mathrm{i}V, [\mathrm{i}\partial_x^2, \mathrm{i}V]] &= -\mathrm{i}[\langle V\rangle_0, [\langle 1\rangle_2, \langle V\rangle_0]] = 2\mathrm{i}\left\langle (\partial_x V)^2\right\rangle_0, \\
[\mathrm{i}\partial_x^2, [\mathrm{i}\partial_x^2, \mathrm{i}V]] &= -\mathrm{i}[\langle 1\rangle_2, [\langle 1\rangle_2, \langle V\rangle_0]] = \mathrm{i}\left\langle \partial_x^4 V\right\rangle_0 - 4\mathrm{i}\left\langle \partial_x^2 V\right\rangle_2.
\end{aligned}
$$

Straightforward discretisations of these operators preserve the symmetries that are crucial for preserving unitarity.

2.2 The expansion for the Schrödinger equation

Using the approach introduced in the previous section, commutators in the Magnus expansion, $\Theta(h) = \sum_{k=1}^{\infty} \Theta^{[k]}(h)$, can be expanded in terms of anti-commutators: $\Theta(h) = \sum_{k=0}^{\infty} \mathrm{i}^{k+1}\langle f_k\rangle_k$. Since $A(t) = \mathrm{i}\partial_x^2 - \mathrm{i}V(t) = \mathrm{i}\langle 1\rangle_2 - \mathrm{i}\langle V(t)\rangle_0$, the first term of the Magnus expansion is the integral $\Theta^{[1]}(h) = \int_0^h A(\zeta)\,\mathrm{d}\zeta$,

$$
\Theta^{[1]}(h) = \mathrm{i}h\langle 1\rangle_2 - \mathrm{i}\int_0^h \langle V(\zeta)\rangle_0\,\mathrm{d}\zeta = \mathrm{i}h\langle 1\rangle_2 - \mathrm{i}\left\langle \int_0^h V(\zeta)\,\mathrm{d}\zeta\right\rangle_0. \tag{2.7}
$$

Note that the integrals here are in time, while differential operators are in space. Along with linearity of the brackets and integrals, this observation allows us to exchange brackets and integrals.

The first non-trivial term. $\Theta^{[2]}(h)$, is simplified as

$$\begin{aligned}\Theta^{[2]}(h) &= -\tfrac{1}{2}\int_0^h\int_0^\zeta [A(\xi), A(\zeta)]\,\mathrm{d}\xi\,\mathrm{d}\zeta \\ &= -\tfrac{1}{2}\int_0^h\int_0^\zeta [\mathrm{i}\,\langle 1\rangle_2 - \mathrm{i}\,\langle V(\xi)\rangle_0\,,\ \mathrm{i}\,\langle 1\rangle_2 - \mathrm{i}\,\langle V(\zeta)\rangle_0]\,\mathrm{d}\xi\,\mathrm{d}\zeta \\ &= -\tfrac{1}{2}\int_0^h\int_0^\zeta [\langle 1\rangle_2\,, \langle V(\zeta)\rangle_0] + [\langle V(\xi)\rangle_0\,, \langle 1\rangle_2]\,\mathrm{d}\xi\,\mathrm{d}\zeta \\ &= -\left(\int_0^h \zeta\,\langle \partial_x V(\zeta)\rangle_1\,\mathrm{d}\zeta - \int_0^h\int_0^\zeta \langle \partial_x V(\xi)\rangle_1\,\mathrm{d}\xi\,\mathrm{d}\zeta\right) \\ &= -\left\langle \int_0^h \zeta(\partial_x V(\zeta))\,\mathrm{d}\zeta - \int_0^h\int_0^\zeta (\partial_x V(\xi))\,\mathrm{d}\xi\,\mathrm{d}\zeta \right\rangle_1 .\end{aligned} \tag{2.8}$$

Higher order terms. Similarly, using (2.6), we can simplify higher nested commutators in the Magnus expansion. For instance,

$$\Theta^{[3,1]}(h) = \frac{1}{12}\int_0^h \left[\int_0^{\xi_1} A(\xi_2)d\xi_2, \left[\int_0^{\xi_1} A(\xi_2)d\xi_2, A(\xi_1)\right]\right] d\xi_1,$$

which occurs as a part of $\Theta^{[3]}(h)$, is simplified to

$$\begin{aligned}\Theta^{[3,1]}(h) = \quad & \tfrac{1}{3}\mathrm{i}\left\langle \int_0^h \zeta^2(\partial_x^2 V(\zeta))\,\mathrm{d}\zeta - \int_0^h \zeta\int_0^\zeta (\partial_x^2 V(\xi))\,\mathrm{d}\xi\,\mathrm{d}\zeta \right\rangle_2 \\ & + \tfrac{1}{6}\mathrm{i}\left\langle \int_0^h \zeta(\partial_x V(\zeta))\int_0^\zeta (\partial_x V(\xi))\,\mathrm{d}\xi\,\mathrm{d}\zeta - \int_0^h \left(\int_0^\zeta (\partial_x V(\xi))\,\mathrm{d}\xi\right)^2 \mathrm{d}\zeta \right\rangle_0 \\ & - \tfrac{1}{12}\mathrm{i}\left\langle \int_0^h \zeta^2(\partial_x^4 V(\zeta))\,\mathrm{d}\zeta - \int_0^h \zeta\int_0^\zeta (\partial_x^4 V(\xi))\,\mathrm{d}\xi\,\mathrm{d}\zeta \right\rangle_0 .\end{aligned} \tag{2.9}$$

2.3 Simplification of integrals

After simplifying terms in the Magnus expansion we arrive at expressions such as (2.8) and (2.9), where each integral is of the form

$$I_{\mathcal{S},\boldsymbol{f}}(h) = \int_{\mathcal{S}} \boldsymbol{f}(\boldsymbol{\xi})\,\mathrm{d}\boldsymbol{\xi},$$

where $\boldsymbol{f}(\boldsymbol{\xi}) = \prod_{j=1}^s f_j(\xi_j)$ for some function f_j, and $\mathcal{S}$ is an s-dimensional polytope of the special form,

$$\mathcal{S} = \{\boldsymbol{\xi} \in \mathbb{R}^s \ : \ \xi_1 \in [0,h], \quad \xi_l \in [0,\xi_{m_l}], \quad l = 2,3,\ldots,s\}, \tag{2.10}$$

where $m_l \in \{1,2,\ldots,l-1\}$, $l=2,3,\ldots,s$. For details about the types of polytopes of integration appearing in the Magnus expansion see (Iserles et al. 2000).

The special form of these polytopes and the integrands obtained after expanding the commutators, allows us to simplify the terms of the Magnus expansion further. Integration by parts leads us to the following identities:

$$\int_0^h f_1(\xi_1)\left(\int_0^{\xi_1} f_2(\xi_2)d\xi_2\right)d\xi_1 = \int_0^h f_2(\xi_1)\left(\int_{\xi_1}^h f_1(\xi_2)d\xi_2\right)d\xi_1, \tag{2.11}$$

$$\int_0^h f_1(\xi_1)d\xi_1\left(\int_0^{\xi_1} f_2(\xi_2)d\xi_2\right)\left(\int_0^{\xi_1} f_3(\xi_3)d\xi_3\right)d\xi_1 \tag{2.12}$$

$$= \int_0^h\left(\int_{\xi_3}^h f_1(\xi_1)d\xi_1\right)\left(f_2(\xi_3)\int_0^{\xi_3} f_3(\xi_2)d\xi_2 + f_3(\xi_3)\int_0^{\xi_3} f_2(\xi_2)d\xi_2\right)d\xi_3.$$

In our simplifications, (2.8) and (2.9), we have already encountered integrals over a triangle such as $\int_0^h\int_0^\zeta(\partial_x V(\xi))\,\mathrm{d}\xi\,\mathrm{d}\zeta$ and $\int_0^h\zeta\int_0^\zeta(\partial_x^2 V(\xi))\,\mathrm{d}\xi\,\mathrm{d}\zeta$. We can reduce these to integrations over a line by applying the first identity with $f_1(\xi_1)=1$, $f_2(\xi_2)=\partial_x V(\xi_2)$ and $f_1(\xi_1)=\xi_1$, $f_2(\xi_2)=\partial_x V(\xi_2)$, respectively. Integration over the pyramid in $\int_0^h\left(\int_0^\zeta(\partial_x V(\xi))\,\mathrm{d}\xi\right)^2\mathrm{d}\zeta$ is similarly reduced using the second identity with $f_1(\xi_1)=1$, $f_2(\xi_2)=\partial_x V(\xi_2)$, $f_3(\xi_3)=\partial_x V(\xi_3)$.

Remark 8 *The use of identities (2.11) and (2.12) is what allows us to reduce the complexity of the integrals in our Magnus expansions. In particular, our order-six method in Subsection 2.8 features integrals over a triangle instead of integrals over four-dimensional polytopes that are typical in the usual order-six Magnus expansions.*

Remark 9 *Although it might be possible to develop general formalism for extending these observations to higher dimensional polytopes appearing in the Magnus expansion, the two identities presented here suffice for all results presented in our work. Deducing similarly useful identities for reduction of nested integrals in any specific high dimensional polytope should also be possible and would be a very helpful result.*

2.4 A proposed Magnus expansion

After simplification of commutators and applications of the integration identities (2.11) and (2.12), the Magnus expansion Θ_3, for instance, reduces to the sum of the following terms,

$$\Theta^{[1]}(h) = \mathrm{i}h\partial_x^2 - \mathrm{i}\int_0^h V(\zeta)\,\mathrm{d}\zeta, \tag{2.13}$$

$$\Theta^{[2]}(h) = -2\left\langle\int_0^h\left(\zeta-\tfrac{h}{2}\right)(\partial_x V(\zeta))\,\mathrm{d}\zeta\right\rangle_1, \tag{2.14}$$

$$\Theta^{[3,1]}(h) = -\tfrac{1}{6}\mathrm{i}\int_0^h\int_0^\zeta (2h-3\zeta)\,(\partial_x V(\zeta))(\partial_x V(\xi))\,\mathrm{d}\xi\,\mathrm{d}\zeta \\ -\tfrac{1}{6}\mathrm{i}\left\langle \int_0^h \left(h^2-3\zeta^2\right)(\partial_x^2 V(\zeta))\,\mathrm{d}\zeta \right\rangle_2, \tag{2.15}$$

$$\Theta^{[3,2]}(h) = \tfrac{1}{2}\mathrm{i}\int_0^h\int_0^\zeta (\zeta-2\xi)\,(\partial_x V(\zeta))(\partial_x V(\xi))\,\mathrm{d}\xi\,\mathrm{d}\zeta \\ +\tfrac{1}{2}\mathrm{i}\left\langle \int_0^h \left(h^2-4h\zeta+3\zeta^2\right)(\partial_x^2 V(\zeta))\,\mathrm{d}\zeta \right\rangle_2, \tag{2.16}$$

where $\Theta^{[3,2]}(h)$ refers to the second part of $\Theta^{[3]}$. Here and in the sequel we prefer to express $\langle f\rangle_0$ as f and $\langle 1\rangle_2$ as ∂_x^2 to avoid an excessively pedantic and longwinded notation.

Remark 10 *The term $\Theta^{[2]}(h) = -2\left\langle \int_0^h \left(\zeta-\frac{h}{2}\right)(\partial_x V(\zeta))\,\mathrm{d}\zeta\right\rangle_1$ might seem to be $\mathcal{O}\left(h^2\right)$ at first sight. A closer look at the special form of the integrand, however, shows that the term is, in fact, $\mathcal{O}\left(h^3\right)$. To observe this, consider $V(\zeta)$ expanded about 0, so that $V(\zeta) = V(0)+\sum_{k=1}^\infty \zeta^k V^{(k)}(0)/k!$. Note that the h^2 term $\int_0^h\left(\zeta-\frac{h}{2}\right)(\partial_x V(0))\,\mathrm{d}\zeta$ vanishes. This is consistent with Remark 1. Similar care has to be exercised throughout the simplifications while analysing size. We refer the reader to (Iserles et al. 2000) for a more general analysis of such gains of powers of h, which occurs in specific terms of the Magnus expansion due to their structure.*

2.5 Simplifying notation

The algebraic workings become increasingly convoluted once we start dealing with larger nested commutators and integrals. Here it becomes helpful to introduce a notation for the integrals on the line,

$$\mu_{j,k}(h) = \int_0^h \tilde{B}_j^k(h,\zeta)V(\zeta)\,\mathrm{d}\zeta, \tag{2.17}$$

and integrals over a triangle,

$$\Lambda\left[f\right]_{a,b}(h) = \int_0^h\int_0^\zeta f(h,\zeta,\xi)\left[\partial_x^a V(\zeta)\right]\left[\partial_x^b V(\xi)\right]\,\mathrm{d}\xi\,\mathrm{d}\zeta, \tag{2.18}$$

where $\tilde{B}$ is a rescaling of Bernoulli polynomials (Abramowitz & Stegun 1964, Lehmer 1988),

$$\tilde{B}_j(h,\zeta) = h^j B_j\left(\zeta/h\right).$$

2.6 Fourth order Magnus expansions

With this new notation in place, the Magnus expansions $\Theta_2(h)$ and $\Theta_3(h)$ can be presented more concisely,

$$\Theta_2(h) = \overbrace{\mathrm{i}h\partial_x^2 - \mathrm{i}\mu_{0,0}(h)}^{\mathcal{O}(h)} - \overbrace{2\left\langle \partial_x \mu_{1,1}(h)\right\rangle_1}^{\mathcal{O}\left(h^3\right)}, \tag{2.19}$$

$$\Theta_3(h) = \Theta_2(h) + \overbrace{\mathrm{i}\Lambda\left[\psi\right]_{1,1}(h) + 2\mathrm{i}\left\langle \partial_x^2 \mu_{2,1}(h)\right\rangle_2}^{\mathcal{O}\left(h^4\right)}, \tag{2.20}$$

where

$$\psi(h,\zeta,\xi) = \zeta - \xi - \tfrac{h}{3}.$$

Due to Remark 1, the error in these Magnus expansions can be expanded solely in odd powers of h since they are based on the power-truncated Magnus expansions of (Iserles et al. 2000). Consequently, both of these expansions are fourth-order.

Remark 11 *Since the* jth *rescaled Bernoulli polynomial scales as* $\mathcal{O}\left(h^j\right)$*, we expect* $\mu_{j,k}(h) = \mathcal{O}\left(h^{jk+1}\right)$ *. Since the integral of the Bernoulli polynomials vanishes,*

$$\int_0^h B_j(h,\zeta)\,\mathrm{d}\zeta = 0, \tag{2.21}$$

however, the term $\mu_{j,1}(h)$ *gains an extra power of* h *and is* $\mathcal{O}\left(h^{j+2}\right)$ *.*

In general, for a polynomial $p_n(h,\zeta,\xi)$ *featuring* only *degree-n terms in* h,ζ *and* ξ*, the linear (integral) functional (2.18) is* $\mathcal{O}\left(h^{n+2}\right)$*. However, the integral of* ψ *over the triangle vanishes,*

$$\int_0^h \int_0^\zeta \psi(h,\zeta,\xi)\,\mathrm{d}\xi\,\mathrm{d}\zeta = 0, \tag{2.22}$$

lending an extra power of h *to terms featuring* $\Lambda\left[\psi\right]_{a,b}(h)$*.*

2.7 Sixth order Magnus expansion

Arbitrarily high order Magnus expansions with simplified commutators can be derived by following the procedure described in the preceding sections. The order six expansion, $\Theta_4(h)$, for instance, is

$$\begin{aligned}
\Theta_4(h) = {}& \overbrace{\mathrm{i}h\partial_x^2 - \mathrm{i}\mu_{0,0}(h)}^{\mathcal{O}(h)} - \overbrace{2\left\langle \partial_x \mu_{1,1}(h)\right\rangle_1}^{\mathcal{O}\left(h^3\right)} + \overbrace{\mathrm{i}\Lambda\left[\psi\right]_{1,1}(h) + 2\mathrm{i}\left\langle \partial_x^2 \mu_{2,1}(h)\right\rangle_2}^{\mathcal{O}\left(h^4\right)} \\
&+ \overbrace{\tfrac{1}{6}\left\langle \Lambda\left[\varphi_1\right]_{1,2}(h) + \Lambda\left[\varphi_2\right]_{2,1}(h)\right\rangle_1}^{\mathcal{O}\left(h^4\right)} + \overbrace{\tfrac{1}{6}\left\langle \Lambda\left[\phi_1\right]_{1,2}(h) + \Lambda\left[\phi_2\right]_{2,1}(h)\right\rangle_1}^{\mathcal{O}\left(h^5\right)} \\
&+ \overbrace{\tfrac{4}{3}\left\langle \partial_x^3 \mu_{3,1}(h)\right\rangle_3}^{\mathcal{O}\left(h^5\right)} + \overbrace{\tfrac{1}{4}\mathrm{i}\partial_x^4 \mu_{2,1}(h)}^{\mathcal{O}\left(h^4\right)} = \Theta(h) + \mathcal{O}\left(h^7\right),
\end{aligned} \tag{2.23}$$

where

$$\begin{aligned}\varphi_1(h,\zeta,\xi) &= h^2 - 4h\xi + 2\zeta\xi, \\ \varphi_2(h,\zeta,\xi) &= (h-2\zeta)^2 - 2\zeta\xi, \\ \phi_1(h,\zeta,\xi) &= h^2 - 6h\zeta + 6h\xi + 6\zeta\xi + 3\zeta^2 - 12\xi^2, \\ \phi_2(h,\zeta,\xi) &= h^2 - 6h\zeta + 6h\xi - 6\zeta\xi + 5\zeta^2.\end{aligned} \tag{2.24}$$

Remark 12 *Integrals of ϕ_j vanish over the triangle,*

$$\int_0^h \int_0^\zeta \phi_j(h,\zeta,\xi)\,\mathrm{d}\xi\,\mathrm{d}\zeta = 0, \quad j = 1,2, \tag{2.25}$$

lending an extra power of h to the functionals wherever ϕ_js appear. No similar observation about φ_js can be made and they have been kept separate in (2.23) from the Λ terms featuring ϕ_j simply since they lead to terms of different orders. For purposes of computing, however, they can be combined.

The linearity of the brackets means that $\Theta_4(h)$ can be rewritten in the form,

$$\Theta_4(h) = \theta_0 + \langle\theta_1\rangle_1 + \langle\theta_2\rangle_2 + \langle\theta_3\rangle_3 \tag{2.26}$$

where

$$\begin{aligned}\theta_0 &= -\mathrm{i}\mu_{0,0}(h) + \mathrm{i}\Lambda\,[\psi]_{1,1}(h) + \tfrac{1}{4}\mathrm{i}\partial_x^4\mu_{2,1}(h), \\ \theta_1 &= -2\partial_x\mu_{1,1}(h) + \tfrac{1}{6}\Lambda\,[\varphi_1+\phi_1]_{1,2}(h) + \tfrac{1}{6}\Lambda\,[\varphi_2+\phi_2]_{2,1}(h), \\ \theta_2 &= \mathrm{i}h + 2\mathrm{i}\partial_x^2\mu_{2,1}(h), \\ \theta_3 &= \tfrac{4}{3}\partial_x^3\mu_{3,1}(h).\end{aligned} \tag{2.27}$$

Written in this form, it is clearly evident that Θ_4 is free of nested commutators and is composed of a very small number of anti-commutators. In fact, the number of anti-commutators grows linearly with the order of accuracy. As we see in the following section, this makes a significant difference to the cost of our methods when compared to standard Magnus–Lanczos schemes (which feature nested commutators and consequently a cost that grows exponentially with order).

Remark 13 *Note that, for potentials of the form $V(x,t) = V_0(x) + f(t)x$, the terms involving $\mu_{1,1}(h), \mu_{2,1}(h)$ and $\mu_{3,1}(h)$ all vanish.*

3 Implementation

In the previous section we proposed the Magnus expansion with simplified commutators. The next step consists in numerically approximating the exponential of this expansion (1.5), which is a challenging problem itself. We will show how Lanczos iterations can be much cheaper when combined with the proposed versions of the Magnus expansion. In this section we present some details of implementation and highlight some crucial features of our schemes.

3.1 Spatial discretisation.

In principle, our methods can be combined with any spatial discretisation strategy, provided the discretisation of ∂_x^n is symmetric for even n and skew-symmetric for odd n. Here we resort to spectral collocation due to its high accuracy. Having imposed periodic boundaries on I, we use equispaced grids with N points. Since we work with values at the grid points, multiplication by the function V (or, in general, function f) is discretised as an $N \times N$ diagonal matrix $\mathcal{D}_V$ (or $\mathcal{D}_f$) with values of V (or f) at the grid points along the diagonal. The differentiation matrices $\mathcal{K}_k$ are symmetric for even k and skew-symmetric for odd k, just as we have assumed throughout. Additionally, spectral collocation results in $\mathcal{K}_k$ being an $N \times N$ circulant. Consequently, it is diagonalisable via Fourier transform,

$$\mathcal{K}_k = \mathcal{F}^{-1}\mathcal{D}_{c_k}\mathcal{F},$$

where c_k is the symbol of $\mathcal{K}_k$ and $\mathcal{F}$ is the $N \times N$ Fourier transform matrix.

Since $\|\mathcal{D}_f\|_2 \le \|f\|_\infty$, the matrix $\mathcal{D}_f$ does not scale with N. On the other hand, it can be verified that $\mathcal{K}_k$ scales as N^k. As previously noted in (2.5), the operator $\langle f\rangle_k$ is discretised as $\frac{1}{2}\left(\mathcal{D}_f\mathcal{K}_k + \mathcal{K}_k\mathcal{D}_f\right)$. Consequently, upon discretisation, $\langle f\rangle_k$ also scales as N^k. We write $\mathcal{K}_k = \mathcal{O}\left(N^k\right)$ and, abusing notation somewhat, $\langle f\rangle_k = \mathcal{O}\left(N^k\right)$.

The order-four Magnus expansion $\Theta_2(h)$, with a local error $\mathcal{O}\left(h^5\right)$, discretises to the form

$$\Theta_2(h) \rightsquigarrow -\mathrm{i}\mathcal{D}_{\mu_{0,0}(h)} - \left(\mathcal{D}_{\partial_x\mu_{1,1}(h)}\mathcal{K}_1 + \mathcal{K}_1\mathcal{D}_{\partial_x\mu_{1,1}(h)}\right) + \mathrm{i}h\mathcal{K}_2, \tag{3.1}$$

while the discretisation of the order-six Magnus expansion $\Theta_4(h)$ with local error $\mathcal{O}\left(h^7\right)$ is

$$\Theta_4(h) \rightsquigarrow \mathcal{D}_{\theta_0} + \tfrac{1}{2}\left(\mathcal{D}_{\theta_1}\mathcal{K}_1 + \mathcal{K}_1\mathcal{D}_{\theta_1}\right) + \tfrac{1}{2}\left(\mathcal{D}_{\theta_2}\mathcal{K}_2 + \mathcal{K}_2\mathcal{D}_{\theta_2}\right) + \tfrac{1}{2}\left(\mathcal{D}_{\theta_3}\mathcal{K}_3 + \mathcal{K}_3\mathcal{D}_{\theta_3}\right). \tag{3.2}$$

3.2 Evaluation of integrals and derivatives of the potential.

Before we implement (3.1) or (3.2) in a practical algorithm, however, we are still left with the task of approximating functions such as $\mu_{0,0}(h)$, $\partial_x\mu_{1,1}(h)$, $\Lambda\left[\psi\right]_{1,1}(h)$ and $\Lambda\left[\varphi_1 + \phi_1\right]_{1,2}(h)$ at the grid points, which are hidden in θ_i in the case of Θ_4. These feature both integrals and derivatives of the potential. In some cases, it might be possible to evaluate some or all of these analytically. In other cases, however, these can be approximated by a combination of quadrature methods and finite difference differentiation[2].

We note that since the derivatives and the integrals are in space and time, respectively, they can be exchanged. Thus the optimal strategy might involve evaluating derivatives first in some cases and integrals first in others. The optimal strategy could also depend on the relative resolutions of temporal and spatial grids. A more challenging scenario is when the temporal grid is coarser than the spatial grid, $h = (\Delta x)^\sigma$

[2]Here we suggest finite differences instead of spectral collocation since the potential is usually less oscillatory and more easily resolved than the wave function. Moreover, as we see shortly, lower degrees of accuracy are required in some cases, allowing us to reduce costs.

for some $0 < \sigma \leq 1$ (in other words, we consider larger time steps). For the sake of simplicity, we follow a fixed strategy of evaluating the derivatives of the potential first.

Derivatives. The various derivatives of V that we need to approximate here, $\partial_x V$, $\partial_x^2 V$, $\partial_x^3 V$ and $\partial_x^4 V$, require differentiation to different degrees of accuracy – by tailoring this accuracy to the $\mathcal{O}\left(h^7\right)$ accuracy of our Magnus expansion we can achieve the required accuracy at a low cost.

Consider $\partial_x \mu_{1,1}(h)$. Due to (2.21), $\int_0^h \tilde{B}_1(h,\zeta) f(\zeta)\,\mathrm{d}\zeta$ is $\mathcal{O}\left(h^3\right)$ for any f. Let $\mathcal{K}_k^{\mathrm{FD},n}$ be the finite difference differentiation matrix approximating ∂_x^k up to an error of $(\Delta x)^n$. This accuracy, expressed in powers of h, naturally depends on the relative sizes of h and Δx, the latter of which is assumed to be fixed. For instance, when $h = \Delta x$, it suffices to approximate $\partial_x V$ to an accuracy of $\mathcal{O}\left((\Delta x)^4\right) = \mathcal{O}\left(h^4\right)$ via $\mathcal{K}_1^{\mathrm{FD},4}\boldsymbol{V}$, since the integral $\int_0^h \tilde{B}_1(h,\zeta)\mathcal{K}_1^{\mathrm{FD},4}\boldsymbol{V}(\zeta)\,\mathrm{d}\zeta$ approximates $\partial_x \mu_{1,1}(h)$ to the required accuracy of $\mathcal{O}\left(h^7\right)$. When the time step is larger, say $h = \sqrt{\Delta x}$, the lower accuracy (and lower cost) differentiation matrix $\mathcal{K}_1^{\mathrm{FD},2}$ suffices. A practical method could use $\mathcal{K}_1^{\mathrm{FD},4}$ for $\Delta x \leq h < \sqrt{\Delta x}$ and $\mathcal{K}_1^{\mathrm{FD},2}$ for $\sqrt{\Delta x} \leq h$.

Similar considerations show that we need to approximate $\partial_x^2 V$ to an accuracy of $\mathcal{O}\left(h^3\right)$, $\partial_x^3 V$ to an accuracy of $\mathcal{O}\left(h^2\right)$ and $\partial_x^4 V$ to an accuracy of $\mathcal{O}\left(h^3\right)$.

Quadrature. For the purpose of approximating the integrals, we can resort to a variety of quadrature methods, among which Gauss–Legendre quadratures are the most popular due to their high orders of accuracy. For instance, all these integrals can be approximated to $\mathcal{O}\left(h^7\right)$ accuracy using Gauss–Legendre quadrature at the knots $\tau_k = h(1 + k\sqrt{3/5})/2$, $k = -1, 0, 1$, with the weights $w_k = \frac{5}{18}h, \frac{4}{9}h, \frac{5}{18}h$ (Davis & Rabinowitz 1984). [3]

Approximation of line integrals. Under $\sigma = 1$, the line integrals $\mu_{j,k}(h)$ and their derivatives appearing in Θ_4 can be approximated to $\mathcal{O}\left(h^7\right)$ accuracy by using the weights w_k,

$$\mu_{0,0}(h) \rightsquigarrow w_{-1}\boldsymbol{V}(\tau_{-1}) + w_0\boldsymbol{V}(\tau_0) + w_1\boldsymbol{V}(\tau_1), \tag{3.3}$$

$$\partial_x \mu_{1,1}(h) \rightsquigarrow w_{-1}\tilde{B}_1(h,\tau_{-1})\mathcal{K}_1^{\mathrm{FD},4}\boldsymbol{V}(\tau_{-1}) + w_1\tilde{B}_1(h,\tau_1)\mathcal{K}_1^{\mathrm{FD},4}\boldsymbol{V}(\tau_1), \tag{3.4}$$

$$\begin{aligned}\partial_x^2 \mu_{2,1}(h) \rightsquigarrow\ & w_{-1}\tilde{B}_2(h,\tau_{-1})\mathcal{K}_2^{\mathrm{FD},3}\boldsymbol{V}(\tau_{-1}) \\ & + w_0\tilde{B}_2(h,\tau_0)\mathcal{K}_2^{\mathrm{FD},3}\boldsymbol{V}(\tau_0) + w_1\tilde{B}_2(h,\tau_1)\mathcal{K}_2^{\mathrm{FD},3}\boldsymbol{V}(\tau_1),\end{aligned} \tag{3.5}$$

$$\begin{aligned}\partial_x^3 \mu_{1,3}(h) \rightsquigarrow\ & w_{-1}\tilde{B}_1(h,\tau_{-1})^3\mathcal{K}_3^{\mathrm{FD},2}\boldsymbol{V}(\tau_{-1}) \\ & + w_0\tilde{B}_1(h,\tau_0)^3\mathcal{K}_3^{\mathrm{FD},2}\boldsymbol{V}(\tau_0) + w_1\tilde{B}_1(h,\tau_1)^3\mathcal{K}_3^{\mathrm{FD},2}\boldsymbol{V}(\tau_1),\end{aligned} \tag{3.6}$$

$$\begin{aligned}\partial_x^4 \mu_{2,1}(h) \rightsquigarrow\ & w_{-1}\tilde{B}_2(h,\tau_{-1})\mathcal{K}_4^{\mathrm{FD},3}\boldsymbol{V}(\tau_{-1}) \\ & + w_0\tilde{B}_2(h,\tau_0)\mathcal{K}_4^{\mathrm{FD},3}\boldsymbol{V}(\tau_0) + w_1\tilde{B}_2(h,\tau_1)\mathcal{K}_4^{\mathrm{FD},3}\boldsymbol{V}(\tau_1),\end{aligned} \tag{3.7}$$

where we note that since $\tilde{B}_1(h,\tau_0) = 0$, the τ_0 term does not appear in (3.4). We note that, instead of (3.5) and (3.7), approximating $\mu_{2,1}(h)$ first and then evaluating its derivatives would be cheaper overall. However, as mentioned earlier, we attempt here to provide a simple and clear procedure, not a fully optimised one.

[3] Recall that since these Magnus expansions are odd in h, the $\mathcal{O}\left(h^6\right)$ Gauss-Legendre quadrature automatically becomes $\mathcal{O}\left(h^7\right)$ in this context.

For the order four Magnus expansion, Θ_2, the first two terms (3.3) and (3.4) suffice. However, since we need only $\mathcal{O}\left(h^5\right)$ accuracy, we could do with just two Gauss–Legendre knots.

Approximation of integrals over the triangle. For the integrals over the triangle such as $\Lambda\left[\psi\right]_{1,1}(h)$, the appropriate weights can be found by substituting the interpolant, $\tilde{\boldsymbol{v}}(t) = \sum_{k=-1}^{1} \ell_k(t)\boldsymbol{v}(\tau_k)$, where $\boldsymbol{v}$ is usually a derivative of the potential[4] and where $\ell_k(t)$ are the Lagrange cardinal functions, $\ell_k(\tau_j) = \delta_{j,k}$. Thus we discretise,

$$\begin{aligned}\Lambda\left[f\right]_{a,b}(h) &\rightsquigarrow \int_0^h \int_0^\zeta \sum_{j=-1}^{1}\sum_{k=-1}^{1} f(h,\zeta,\xi)\ell_j(\zeta)\ell_k(\xi)\left[\mathcal{K}_a^{\mathrm{FD},r}\boldsymbol{V}(\tau_j)\right]\left[\mathcal{K}_b^{\mathrm{FD},r}\boldsymbol{V}(\tau_k)\right]\mathrm{d}\xi\,\mathrm{d}\zeta \\ &= \sum_{j=-1}^{1}\sum_{k=-1}^{1} w_{jk}^f\left[\mathcal{K}_a^{\mathrm{FD},r}\boldsymbol{V}(\tau_j)\right]\left[\mathcal{K}_b^{\mathrm{FD},r}\boldsymbol{V}(\tau_k)\right], \qquad (3.8)\end{aligned}$$

where we need to approximate derivatives of V to order r (under the scaling $\sigma = 1$, $r = 3$ suffices for all Λ terms in Θ_4 for $\mathcal{O}\left(h^7\right)$ accuracy), and where w_{jk}^f are the weights specific to f,

$$w_{jk}^f = \int_0^h \int_0^\zeta f(h,\zeta,\xi)\ell_j(\zeta)\ell_k(\xi)\,\mathrm{d}\xi\,\mathrm{d}\zeta.$$

The weights for the functions $\psi, \varphi_1, \varphi_2, \phi_1$ and ϕ_2 that are required for the implementation of an order-six method have been provided in Appendix A. The method for discretising the anti-commutators, as well as a particular recipe for approximating the integrals and derivatives of the potential, is in place.

Remark 14 *Having elaborated on the use of Gauss–Legendre quadratures in developing a specific scheme, we remind the reader that a major advantage of preserving integrals throughout the workings in Section 2 is the flexibility of allowing alternative means for evaluating integrals and derivatives, including the possibility of exact integration and derivation.*

3.3 Approximation of the exponential of a Magnus expansion.

After discretising $\Theta_2(h)$ and $\Theta_4(h)$, we are left with the task of approximating their exponential in (2.3) in order to find the solution

$$\boldsymbol{u}^1 = \exp(\Theta_m(h))\boldsymbol{u}^0.$$

As discussed in Subsection 1.1, Lanczos iterations are a very effective, and perhaps the most popular, means for approximating the exponential of a Magnus expansion. This will be the approach adopted in this paper.

Approximation of the matrix vector product $\exp(\Theta_p)\boldsymbol{u}$ via Lanczos iterations requires the evaluation of $\Theta_p\boldsymbol{v}$ in each Lanczos iteration. So long as the number of steps is reasonably small, the cost is dominated by the cost of evaluating $\Theta_p\boldsymbol{v}$.

[4]For instance, $\boldsymbol{v}(t) = \mathcal{K}_1^{\mathrm{FD},3}\boldsymbol{V}(t)$ suffices in the approximation of $\Lambda\left[\psi\right]_{1,1}(h)$.

Standard Magnus expansions feature nested commutators. In the method presented in Subsection 1.1, Θ_p features commutators nested to $p-1$ levels. For a commutator C_p that is nested to p levels, the cost of evaluating the matrix-vector product $C_p\boldsymbol{v}$ grows exponentially with p. Consequently, the cost of $\Theta_p\boldsymbol{v}$ in standard Magnus–Lanczos schemes grows exponentially with the order of the scheme. In contrast our proposed Magnus expansions feature a linearly growing number of non-nested terms. As evident from (3.2), for instance our $\mathcal{O}\left(h^7\right)$ method $\Theta_4(h)$ is comprised of a small number of terms. The approximation of $\Theta_4\boldsymbol{v}$ in each Lanczos iterations requires us to compute

$$\begin{aligned}
\Theta_4(h)\boldsymbol{v} &= \mathcal{D}_{\theta_0}\boldsymbol{v} + \tfrac{1}{2}\left(\mathcal{D}_{\theta_1}\mathcal{K}_1 + \mathcal{K}_1\mathcal{D}_{\theta_1}\right)\boldsymbol{v} + \tfrac{1}{2}\left(\mathcal{D}_{\theta_2}\mathcal{K}_2 + \mathcal{K}_2\mathcal{D}_{\theta_2}\right)\boldsymbol{v} + \tfrac{1}{2}\left(\mathcal{D}_{\theta_3}\mathcal{K}_3 + \mathcal{K}_3\mathcal{D}_{\theta_3}\right)\boldsymbol{v} \\
&= \mathcal{D}_{\theta_0}\boldsymbol{v} + \tfrac{1}{2}\left(\mathcal{D}_{\theta_1}\mathcal{F}^{-1}\mathcal{D}_{c_1}\mathcal{F} + \mathcal{F}^{-1}\mathcal{D}_{c_1}\mathcal{F}\mathcal{D}_{\theta_1}\right)\boldsymbol{v} \\
&\quad + \tfrac{1}{2}\left(\mathcal{D}_{\theta_2}\mathcal{F}^{-1}\mathcal{D}_{c_2}\mathcal{F} + \mathcal{F}^{-1}\mathcal{D}_{c_2}\mathcal{F}\mathcal{D}_{\theta_2}\right)\boldsymbol{v} + \tfrac{1}{2}\left(\mathcal{D}_{\theta_3}\mathcal{F}^{-1}\mathcal{D}_{c_3}\mathcal{F} + \mathcal{F}^{-1}\mathcal{D}_{c_3}\mathcal{F}\mathcal{D}_{\theta_3}\right)\boldsymbol{v} \\
&= \mathcal{D}_{\theta_0}\boldsymbol{v} + \tfrac{1}{2}\left(\sum_{j=1}^{3}\mathcal{D}_{\theta_j}\mathcal{F}^{-1}\mathcal{D}_{c_j}\right)\mathcal{F}\boldsymbol{v} + \tfrac{1}{2}\mathcal{F}^{-1}\left(\sum_{j=1}^{3}\mathcal{D}_{c_j}\mathcal{F}\mathcal{D}_{\theta_j}\boldsymbol{v}\right),
\end{aligned} \tag{3.9}$$

which requires merely eight FFTs.

3.4 Unitarity, norm preservation and stability

Note that our Magnus expansions (2.19) and (2.26) are of the form $\sum_{k=0}^{\infty} \mathrm{i}^{k+1}\left\langle\theta_k\right\rangle_k$ for some θ_k, and it can be seen that each term $\mathrm{i}^{k+1}\left\langle\theta_k\right\rangle_k$ discretises to a skew-Hermitian form in (3.1) and (3.2), respectively. The Magnus expansion, developed in this way preserves skew-Hermiticity and its exponential therefore preserves unitarity. As mentioned in Subsection 1.1, this is consistent with a central feature of quantum mechanics. Additionally, since the exponential is unitary,

$$\|\boldsymbol{u}^1\|_2 = \|\exp\left(\Theta_m(h)\right)\boldsymbol{u}^0\|_2 = \|\boldsymbol{u}^0\|_2,$$

and the norm of $\boldsymbol{u}$ is preserved. Consequently, unitarity guarantees stability of our schemes under any scaling of h and Δx.

4 Numerical Examples

The initial condition for our numerical experiments is a Gaussian wavepacket

$$u_0(x) = (\delta\pi)^{-1/4}\exp\left((-(x-x_0)^2)/(2\delta)\right), \quad x_0 = -2.5, \quad \delta = 0.2,$$

sitting in the left well of a double well potential,

$$V_{\mathrm{D}}(x) = x^4 - 20x^2.$$

We take $[-10, 10]$ as our spatial domain and $[0, 5]$ as our temporal domain. When we allow the wave function to evolve under V_{D}, it remains largely confined to the left well at the final time, $T = 5$ (see Figure 4.2, top left). Superimposing a time dependent excitation to the potential, we are able to exert control on the wave function. In

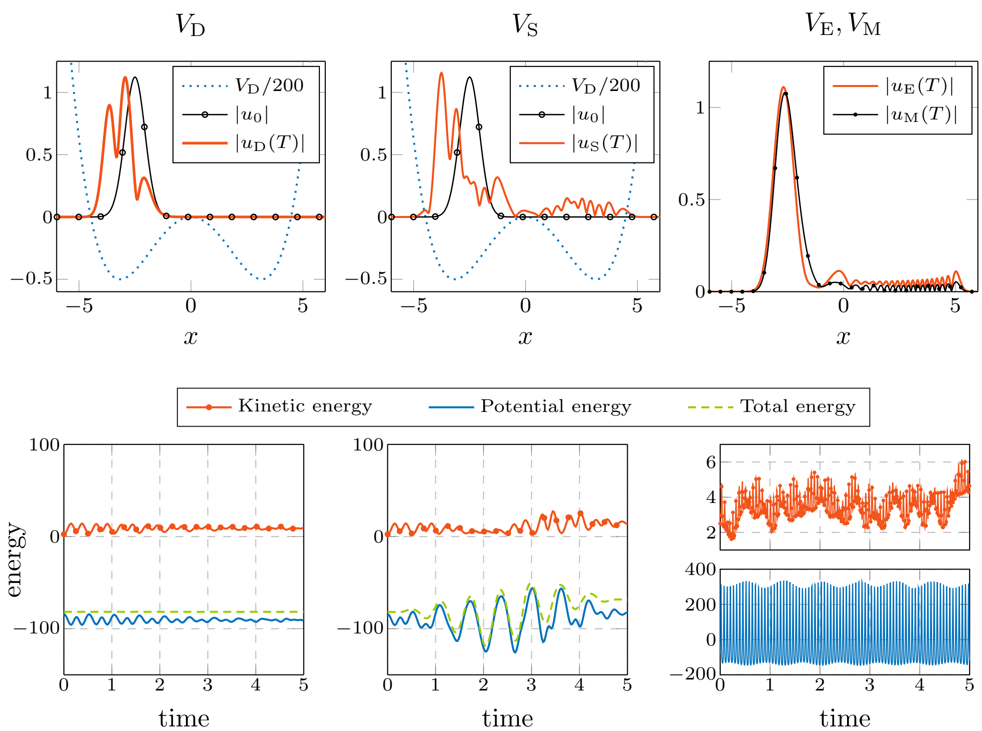

Figure 4.2: **[top row]** The initial condition u_0 evolves to u_{D} under the influence of V_{D} (left), to u_{S} under V_{S} (centre) and to u_{E} and u_{M} under V_{E} and V_{M}, respectively (right). The potential V_{D} is scaled down for ease of presentation. **[bottom row]** Corresponding evolution of energies.

Figure 4.2 (top centre and right) we show the influence of two excitations of the form $f(t)\,x$ with the choices $f(t) = 10\,\mathrm{S}_{10,T}(t)$ and $f(t) = -25\,\mathrm{E}_{100}(t)$, where

$$\mathrm{S}_{\omega,T}(t) = \sin(\pi t/T)^2 \sin(\omega t), \quad \mathrm{E}_{\omega,T}(t) = \exp(2\sin(\omega t)) - 1.$$

The effective time-varying potentials in these cases are

$$V_{\mathrm{S}}(x,t) = V_{\mathrm{D}}(x) + 10\,\mathrm{S}_{10,T}(t)x, \quad V_{\mathrm{E}}(x,t) = V_{\mathrm{D}}(x) - 25\,\mathrm{E}_{100}(t)x,$$

respectively. Since the potentials are available in their analytic form, we use analytic derivatives in our implementation. The integrals were approximated via three Gauss–Legendre knots, as outlined in Subsection 3.2. In principle we can also use analytic or asymptotic approximations for the integrals. Potential accuracy advantages of resorting to analytic approximations should become evident by comparing with a higher degree quadrature – for this purpose we also present results using eleven Gauss–Legendre knots.

Mean field approximation (V_{M}). The excitation in V_{S} features a low frequency oscillation at $\omega = 10$, while V_{E} has a higher frequency oscillation, $\omega = 100$. In the limit $\omega \to \infty$ the effect of the potential function can be approximated by a mean field potential[5] and it is worth finding out to what extent this approximation suffices for V_{E}. Since $\int_0^5 \mathrm{E}_{100}(t)\,\mathrm{d}t \approx 6.45083$, the (time-independent) mean field potential is roughly

$$V_{\mathrm{M}}(x) = V_{\mathrm{DW}}(x) - 32.25415x.$$

In Figure 4.2 (top right) it is evident that the mean field solution $u_{\mathrm{M}}(T)$ is not a sufficiently accurate approximation to $u_{\mathrm{E}}(T)$, and at $\omega = 100$ we do require a solution via high-order Magnus based methods.

Magnus–`expm`. In the numerical experiments presented in this section, order four and six traditional Magnus expansions are denoted by M_4 and M_6, respectively, while the corresponding Magnus expansions with simplified commutators are denoted by S_4 and S_6, respectively. The order four methods use two Gauss–Legendre quadrature knots, while order six methods use three knots. All these methods use 180 spatial grid points and are exponentiated using MATLAB's `expm`. We present the errors for these Magnus–`expm` methods in order to study the error inherent in the Magnus expansion separately from the error due to Lanczos iterations.

Higher accuracy quadratures. In a high frequency regime, we encounter more oscillations per time step and three quadrature knots can be inadequate for approximating the integrals adequately. In such cases, we can expect to see a considerable advantage when using analytic integrals, asymptotic approximations or higher accuracy quadratures. This behaviour is exhibited in Figure 4.4, where our order six integral preserving Magnus expansion S_6 is seen to have a much higher accuracy than the standard Magnus expansion M_6, particularly when combined with a higher accuracy approximation to the integrals. In this case, we resort to 11 Gauss–Legendre quadratures, denoted by the postfix G_{11}. Analytic integrals could possibly improve the accuracy further, as could highly oscillatory quadrature methods (Deaño, Huybrechs & Iserles 2018), which can be easily transplanted to this setting.

Magnus–Lanczos. In Figure 4.3 (right) and Figure 4.4 (right) we show the errors in exponentiating the order six Magnus expansions (with and without simplification of commutators) via 50, 20 and 10 Lanczos iterations respectively. The Magnus–Lanczos schemes with n Lanczos iterations are denoted with the postfix L_n. It is evident from these figures that exponentiation of Magnus expansions via Lanczos iterations requires either a larger number of Lanczos iterations or smaller time steps before we achieve the accuracy inherent in the Magnus expansion (i.e. the accuracy of brute force exponentiation, M_6 and S_6).

Figures 4.3 (right) and 4.4 (right) suggest that there is scope for improvement in the efficient exponentiation of Magnus expansions, particularly when it comes to large time steps, which can be crucial for long term integration. In particular, it is worth exploring whether Zassenhaus splittings confer an advantage here.

The convergence of Lanczos approximation to the exponential can be very sensitive to the degree of spatial discretisation. $\mathrm{M}_6\mathrm{L}_{50}\mathrm{H}$, in Figure 4.3 was run using 1024 grid points. Not only are the Lanczos iterations more expensive in this case, but the convergence also occurs much later. In general we need more iterations since the spectral radius of the Magnus expansion is larger (growing quadratically with finer

[5] Effectively the first and trivial Magnus expansion Θ_1.

spatial resolution).

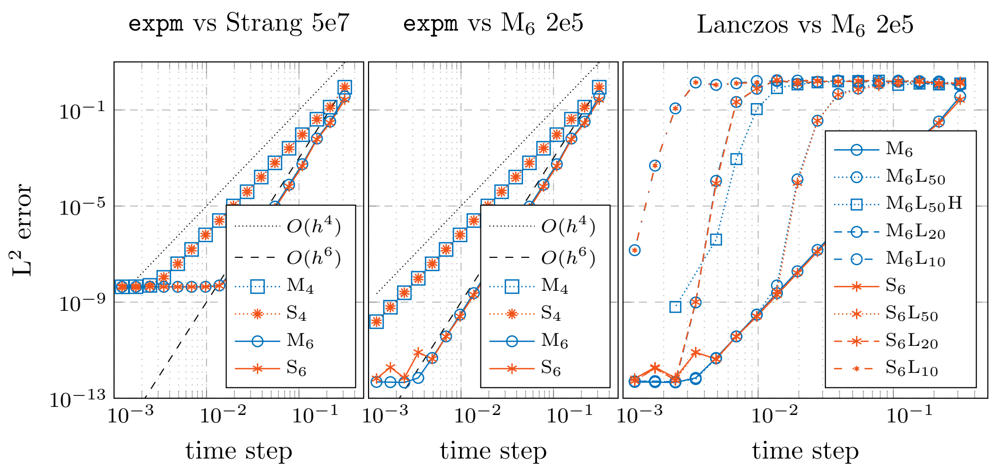

Figure 4.3: [**Low oscillation regime** (V_S)]: When applied to the low oscillatory regime of V_S, the order four and order six Magnus expansions with simplified commutators, S_4 and S_6, have a similar error as the standard Magnus expansions, M_4 and M_6. Not much difference is made in this case (V_S) by considering higher-order quadrature. On the left we use a Strang splitting (exponential midpoint rule) with 1024 grid points and 5×10^7 time steps as a reference. As we can see, the errors saturate around 10^{-8}, which is the accuracy of this reference solution. For the other two plots we use M_6 with 180 grid points and 2×10^5 time steps.

Remark 15 *Note that, for potentials of the form $V(x,t) = V_0(x) + f(t)x$, the terms involving $\mu_{1,1}(h), \mu_{2,1}(h)$ and $\mu_{3,1}(h)$ all vanish. This property, however, has not been exploited in the results presented here.*

5 Conclusions

In this paper we have presented the derivation of integral-preserving Magnus–Lanczos methods with simplified commutators (of arbitrarily high orders) for the computation of the Schrödinger equation featuring time-dependent potentials (1.1) under the atomic scaling, $\hbar = 1$. In particular, we have presented the 4th and 6th order methods ((2.19) and (2.26–2.27), respectively) and analysed their complexity in terms of the number of FFTs required in each Lanczos iteration (see (3.9)).

We find that the number of FFTs is much smaller than for standard Magnus expansions where commutators appear explicitly – our sixth-order method, for instance, requires merely 8 FFTs for each Lanczos iteration. This speedup is evident in numerical experiments (see Figure 4.5, where we find that our method is roughly 7 to 9 times faster than standard Magnus–Lanczos methods). Moreover, the number of FFTs can be shown to grow linearly with the order of the method we seek, so that the 8th order

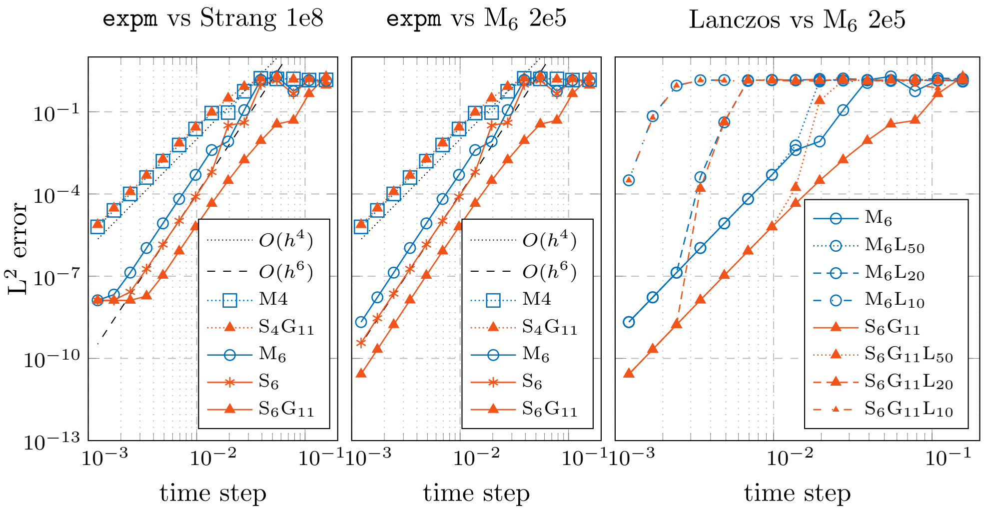

Figure 4.4: [**Highly oscillatory regime** (V_{E})]: In the highly oscillatory regime of V_{E}, we also include the 11 Gauss–Legendre quadrature knots versions of S_4 and S_6, which are labeled with the postfix G_{11}. A significant difference is made in this case (V_{E}) by considering higher-order quadrature. Analytic expressions for integrals could be beneficial in such cases. On the left we use a Strang splitting (exponential midpoint rule) with 512 grid points and 10^8 time steps as a reference. For the other two plots we use M_6 with 180 grid points and 2×10^5 time steps for the reference solution.

method in this class of methods would require 10 FFTs and have a more pronounced speedup over the standard Magnus expansion of order 8.

A concrete example of discretising the integrals via Gauss–Legendre quadrature is also presented in (3.3–3.8). However, as stressed throughout, one of the major advantages of our approach is the flexibility of choosing the method for approximating the integrals at the very last stage. This is likely to prove highly beneficial in the case of highly oscillatory potentials.

To illustrate this advantage, we present a numerical example featuring a highly oscillatory potential, V_{E}, in Section 4. Here we find that our order-six Magnus expansion using three Gauss–Legendres knots is roughly six times more accurate than the standard Magnus expansion. This is improved significantly by resorting to eleven Gauss–Legendre knots, resulting in the accuracy being roughly 80 times higher than the standard Magnus expansion (see Figure 4.4).

5.1 Future work

Approximation of integrals. It should be possible to improve upon the accuracy and cost further by using analytic integrals, asymptotic approximations or specialised highly-oscillatory quadrature.

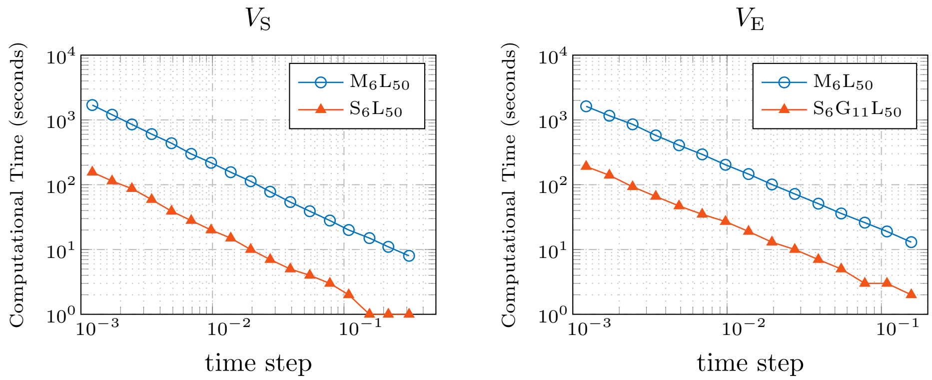

Figure 4.5: **[Computational Time ($V_{\mathrm{S}}, V_{\mathrm{E}}$)]:** The absence of nested commutators in our proposed Magnus expansions results in a significant improvement in computational time of a corresponding Magnus–Lanczos scheme ($\mathrm{S}_6\mathrm{L}_{50}$ is roughly 7 to 9 times faster here than $\mathrm{M}_6\mathrm{L}_{50}$). This effect becomes more pronounced once we consider higher-order Magnus expansions.

Larger time steps. We remind the reader that the accuracy inherent in the Magnus expansion (as evident via direct exponentiation) is only reflected in the Magnus–Lanczos methods when combined with a sufficient number of Lanczos iterations (see Figure 4.3 (right) and Figure 4.4 (right)). This is not much of a constraint when we need high accuracy but can afford to work with moderately large to small time steps since a reasonable number of Lanczos iterations suffices in this regime. However, in applications where cost constraints trump accuracy requirements and necessitate significantly larger time steps, the number of Lanczos iterations required might become a concern.

In these regimes, it might be preferable to resort to Chebyshev expansions which have been found to be more effective than Lanczos iterations for larger time steps. Asymptotic splittings such as symmetric Zassenhaus splittings are also likely to prove effective. This is because the number of Lanczos iterations is dominated by the (spectral) size of the $\mathcal{O}(h)$ terms such as $\mathrm{i}h\partial_x^2$ and $-\mathrm{i}\mu_{0,0}(h)$ that arise from $\Theta^{[1]}(h)$, while the cost of each Lanczos iteration in a high order expansion is dominated by the trailing terms arising from $\Theta^{[k]}(h), k > 1$. Symmetric Zassenhaus splittings can effectively separate terms by powers of h and are likely to prove effective in decoupling these factors affecting the cost and accuracy of Lanczos iterations.

Other equations. It might be possible to extend some of these techniques for other equations of quantum mechanics and certain linear parabolic equations where Magnus expansions are employed. In particular, it might be possible to simplify commutators[6] which could reduce cost, while preserving integrals in the case of highly oscillatory forcing could increase accuracy.

[6]In applications where there is no need to preserve skew-Hermiticity, it would suffice to expand in terms of $f(x)\partial_x^k$ instead of working with $\langle f\rangle_k$ (see Section 2).

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A Quadrature weights

Once we have the values of V at the set of knots K, the integrals over the triangle can be approximated via (3.8),

$$\Lambda\left[f\right]_{a,b}(h) \rightsquigarrow \sum_{j\in K}\sum_{k\in K} w^f_{jk}\left[\mathcal{K}_a \boldsymbol{V}(\tau_j)\right]\left[\mathcal{K}_b \boldsymbol{V}(\tau_k)\right] \text{ or } \sum_{j\in K}\sum_{k\in K} w^f_{jk}\left[\boldsymbol{\partial^a_x V}(\tau_j)\right]\left[\boldsymbol{\partial^b_x V}(\tau_k)\right],$$

depending on whether exact derivatives $\partial^a_x V$ and $\partial^b_x V$ are available or not (in the latter case we resort to numerical differentiation via $\mathcal{K}_a$ and $\mathcal{K}_b$). As usual, boldface denotes a vector of values resulting from spatial discretisation. The weights required for three Gauss–Legendre quadrature knots for the functions $\psi, \varphi_1, \varphi_2, \phi_1$ and ϕ_2 are

$$\begin{aligned}
w^{\psi} &= \left(\frac{h}{6}\right)^3 \left\{ \frac{1}{63}\begin{pmatrix} -139 & 26 & 239 \\ 26 & -304 & 26 \\ 239 & 26 & -139 \end{pmatrix} + 5\sqrt{\frac{3}{5}}\begin{pmatrix} 0 & 0 & -1 \\ 0 & 0 & 0 \\ 1 & 0 & 0 \end{pmatrix} \right\}, \\
w^{\varphi_1} &= \frac{2}{7}\left(\frac{h}{6}\right)^4 \left\{ \begin{pmatrix} -11 & -62 & 136 \\ 190 & -128 & 190 \\ 136 & -62 & -11 \end{pmatrix} + \sqrt{\frac{3}{5}}\begin{pmatrix} 175 & 58 & -170 \\ 222 & 0 & -222 \\ 170 & -58 & -175 \end{pmatrix} \right\}, \\
w^{\varphi_2} &= 2\left(\frac{h}{6}\right)^4 \left\{ \begin{pmatrix} -5 & -14 & 10 \\ -2 & -32 & -2 \\ 10 & -14 & -5 \end{pmatrix} + \frac{1}{7}\sqrt{\frac{3}{5}}\begin{pmatrix} 145 & 134 & -90 \\ -46 & 0 & 46 \\ 90 & -134 & -145 \end{pmatrix} \right\}, \\
w^{\phi_1} &= \frac{2}{7}\left(\frac{h}{6}\right)^4 \left\{ \begin{pmatrix} -17 & 160 & -143 \\ -92 & 184 & -92 \\ -143 & 160 & -17 \end{pmatrix} + 6\sqrt{\frac{3}{5}}\begin{pmatrix} 25 & -34 & 30 \\ -16 & 0 & 16 \\ -30 & -34 & -25 \end{pmatrix} \right\}, \\
w^{\phi_2} &= \left(\frac{h}{6}\right)^4 \left\{ 6\begin{pmatrix} 3 & 0 & -3 \\ -4 & 8 & -4 \\ -3 & 0 & 3 \end{pmatrix} + \frac{4}{7}\sqrt{\frac{3}{5}}\begin{pmatrix} 25 & -2 & 40 \\ -48 & 0 & 48 \\ -40 & 2 & -25 \end{pmatrix} \right\}.
\end{aligned}$$