On the discretisation of the semiclassical Schrödinger equation with time-dependent potential

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Abstract

The computation of the semiclassical Schrödinger equation featuring time-dependent potentials is of great importance in quantum control of atomic and molecular processes. It presents major challenges because of the presence of a small parameter. Assuming periodic boundary conditions, the standard approach in tackling this problem consists of semi-discretisation with a spectral method, followed by a Magnus expansion. It is typical to discretise in time, thereafter replacing all integrals occurring in the Magnus expansion with quadratures. Following this, an exponential splitting is usually prescribed. In this paper we sketch an alternative strategy where semi-discretisation and approximation of integrals is done at the very end, following an exponential splitting. This approach allows us to consider significantly larger time steps and gives us the flexibility to handle a variety of potentials, inclusive of highly oscillatory potentials. Our analysis commences from the investigation of the free Lie algebra generated by differentiation and by multiplication with the interaction potential. It turns out that this algebra possesses structure that renders it amenable to a very effective form of *asymptotic splitting:* an exponential splitting where consecutive terms are scaled by increasing powers of the small parameter. This leads to methods that attain high spatial and temporal accuracy and whose cost scales like $\mathcal{O}(M \log M)$, where M is the number of degrees of freedom in the discretisation.

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1 Introduction

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

$$\frac{\partial u(x,t)}{\partial t} = \mathrm{i}\varepsilon \frac{\partial^2 u(x,t)}{\partial x^2} + \mathrm{i}\varepsilon^{-1} V(x,t)u(x,t), \quad x \in \mathbb{R},\ t \geq 0, \tag{1.1}$$

where the wavefunction $u = u(x,t)$ is given with an initial condition $u(x,0) = u_0(x)$. Here $0 < \varepsilon \ll 1$ is the *semiclassical* parameter whose small size affects rapid oscillations of solution which cause obvious difficulties with numerical discretisation.

Historically, the time-independent Schrödinger equation (TISE) preceded the development of the time-dependent Schrödinger equation (TDSE). Briggs & Rost (2001) note that Schrödinger considered time dependence in the TDSE as arising from time-dependent potentials, in a classical treatment of the external environment. One of the first applications of the TDSE that Schrödinger considered was to the interaction of an atom with a classical electric field, resulting in a time-dependent potential, $V(t)$. Time-dependent potentials, in this way, can be considered to have a fundamental relevance to the TDSE.

As laser technology matures and ability to manipulate electric and magnetic fields increases, unprecedented quantum control of atomic and molecular systems is becoming possible (Shapiro & Brumer 2003). Numerical solution of TDSE with time-varying potentials should prove particularly relevant in this field.

Consider the evolution of the wave-packet

$$u_0(x) = (\delta\pi)^{-1/4} \exp\left(-\frac{(x-x_0)^2}{2\delta}\right) \tag{1.2}$$

with

$$x_0 = -0.3, \quad \delta = 1.22 \times 10^{-4},$$

sitting in the double-well potential

$$V_0 = -(450x^4 - 100x^2 + 1). \tag{1.3}$$

The negative sign accounts for our convention since traditionally the potential in (1.1) appears with the opposite sign. It is typical to impose periodic boundary conditions in order to resolve spatial oscillations with spectral accuracy. We restrict the domain to $[-1,1]$, imposing periodic boundaries at $x = \pm 1$.

Figure 1.1 shows that the wave-packet $u(T)$ at $T = 0.1$, evolving under the influence of the time-independent double-well potential V_0 alone, does not leave the left well. When we excite the wave-packet with the time-varying potential,

$$L_\omega(x,t) = 2\rho(t)\sin((x-5t)\pi\omega), \tag{1.4}$$

however, we find that we are able to induce part of the wave-packet to move to the second well. Figure 1.1 shows the final wave-packet $u_L(T)$ under the influence of the potential $V(x,t) = V_0 + L_{100}(x,t)$. Here the semiclassical parameter is taken to be

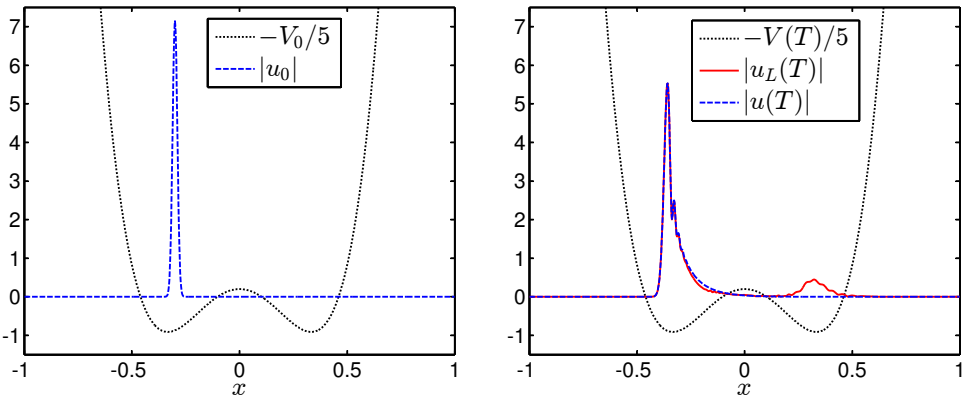
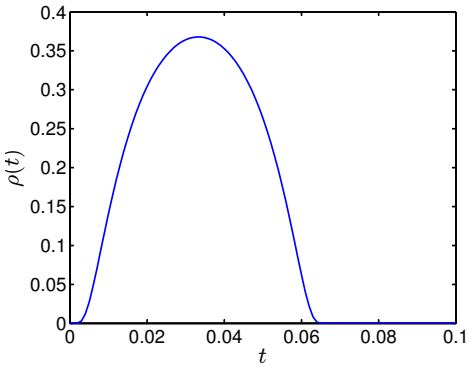


Figure 1.1: (left) initial wave-packet $u(0) = u_0$, (right) final wave-packets at time $T = 1$: $u(T)$ under the influence of V_0 and $u_L(T)$ under the influence of $V_0 + L_{100}(x, t)$. For consistency with physical interpretations we depict the negative potential, scaling it down by a factor of five for ease of illustration.

$\varepsilon = 0.01$ and

$$\rho(t) = \begin{cases} \mathrm{e}^{-\frac{1}{1-(30t-1)^2}} & \text{for } |30t-1| < 1, \\ 0 & \text{otherwise} \end{cases}$$



is a bump function which acts as a smooth envelope simulating the switching on and off of the time-varying component of the potential.

Considering (1.1) to be an evolutionary PDE evolving in a Hilbert space, say $\mathcal{H} = \mathrm{L}_2[-1, 1]$, and suppressing the dependence on x,

$$\partial_t u(t) = \left(\mathrm{i}\varepsilon\partial_x^2 + \mathrm{i}\varepsilon^{-1}V(t)\right) u(t), \ u(0) = u_0, \tag{1.5}$$

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

$$\partial_t u(t) = A(t)u(t), \ u(0) = u_0, \tag{1.6}$$

with $A(t) := \mathrm{i}\varepsilon\partial_x^2 + \mathrm{i}\varepsilon^{-1}V(t)$. 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 corresponding to $\mathfrak{u}(\mathcal{H})$.

Unitary evolution of the wavefunction, $u(t)$, under this flow is a central aspect of quantum mechanics. Preservation of this property under discretisation is a desirable aspect of any numerical method which naturally comes about when working in the appropriate Lie-algebraic framework. Note that unitarity automatically guarantees stability of a consistent numerical scheme.

We impose periodic boundary conditions at ± 1 to allow an effective approximation by spectral methods and 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 $\mathrm{C}_p^\infty([-1,1];\mathbb{R})$ and $\mathrm{C}_p^\infty([-1,1];\mathbb{C})$, respectively, the spaces of real valued and complex valued smooth periodic functions over $[-1,1]$, but our results extend in a straightforward manner to functions of lower smoothness.

Traditionally, the first step in approximating (1.5) is spatial discretisation,

$$\boldsymbol{u}'(t) = \mathrm{i}(\varepsilon\mathcal{K}^2 + \varepsilon^{-1}\mathcal{D}_{V(\cdot,t)})\boldsymbol{u}(t), \qquad t \geq 0, \tag{1.7}$$

where vector $\boldsymbol{u}(t) \in \mathbb{C}^M$ represents an approximation to the solution at time t, $\boldsymbol{u}(0)$ 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. Hochbruck & Lubich (2002) propose solving (1.7) through a Magnus expansion,

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

where $\boldsymbol{\Theta}(h) \in \mathfrak{u}_M(\mathbb{C})$ is a time-dependent $M \times M$ skew-Hermitian matrix obtained as an infinite series $\sum_{k=1}^\infty \boldsymbol{\Theta}^{[k]}(h)$ with each $\boldsymbol{\Theta}^{[k]}(h)$ composed of k nested integrals and commutators of the matrices $\mathrm{i}\varepsilon\mathcal{K}^2$ and $\mathrm{i}\varepsilon^{-1}\mathcal{D}_V$. The authors conclude that $h\|\mathcal{K}\| \leq c$ for some constant c and time step h, suffices for the convergence of the Magnus expansion $\boldsymbol{\Theta}(h)$. In the context of the Schrödinger equation (1.1), this represented a significant improvement over the more stringent convergence requirements of (Iserles & Nørsett 1999, Moan & Niesen 2008) which were obtained under a very general setting.

Jin, Markowich & Sparber (2011) note that the solution of the Schrödinger equation develops oscillations of order $\mathcal{O}\left(\varepsilon^{-1}\right)$, necessitating a large number of degrees of freedom in the spatial discretisation, $M = \mathcal{O}\left(\varepsilon^{-1}\right)$. Since the differentiation matrix $\mathcal{K}$ scales as $\mathcal{O}(M) = \mathcal{O}\left(\varepsilon^{-1}\right)$, the constraint on the time step $h = \mathcal{O}(\varepsilon)$ is considerable. Additionally, $\boldsymbol{\Theta}(h)$ ends up being a large matrix (spectrally as well as in size) which does not posses any favourable structure that could allow an effective approximation of the exponential $\exp(\boldsymbol{\Theta}(h))$.

Splitting methods (Yošida 1990, McLachlan & Quispel 2002, Blanes, Casas & Murua 2006, Lubich 2008, Jin et al. 2011, Faou 2012) can separate some spectrally large but structurally favourable components of the form $\mathrm{i}\varepsilon\mathcal{K}^2$ and $\mathrm{i}\varepsilon^{-1}\mathcal{D}$ from $\boldsymbol{\Theta}(h)$. These components are either diagonal or circulant matrices and are either exponentiated directly or through a couple of Fast Fourier Transforms (FFTs) which can diagonalise the circulant. The remaining parts of $\boldsymbol{\Theta}(h)$ end up being spectrally smaller and may be exponentiated via Lanczos iterations (Gallopoulos & Saad 1992, Hochbruck & Lubich 1997). However, the large size and inconvenient structure of commutators

occurring in $\mathbf{\Theta}(h)$ and its splittings makes this approach suboptimal, particularly once we seek higher-order schemes.

Our narrative develops along a different path: in line with the approach of (Bader, Iserles, Kropielnicka & Singh 2014), the semidiscretisation is deferred to the very last moment. This enables us to take a subtle but powerful advantage of working with undiscretised operators, ∂_x^2 and V, which, when properly applied, simplifies expressions and allows us to lower the complexity of the method.

In contrast to the approach of (Hochbruck & Lubich 2002), we seek a Magnus expansion for the *undiscretised* equation (1.5),

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

where h is a small time step and $\Theta(h) \in \mathfrak{u}(\mathrm{C}_p^\infty([-1,1];\mathbb{C}))$ is a skew-Hermitian *operator* acting on the space of smooth periodic functions over $[-1,1]$. The Magnus expansion $\Theta(h)$, introduced in section 2, features nested integrals of commutators of A.

In Section 3, we introduce simplification rules of the *free Lie algebra* of ∂_x^2 and V, $\mathrm{FLA}\{\partial_x^2, V\}$. We observe that once we work with the undiscretised operators, it is possible to expand commutators and arrive at a commutator-free Magnus expansion for (1.5). The property of *height reduction* summed up in Lemma 1 and Corollary 1 is a clear demonstration of the systematic reduction in complexity which results from working with undiscretised operators.

Terms of the Magnus expansion obtained in this way do not seem to be skew-Hermitian, however. A straightforward discretisation at this stage would result in loss of unitarity and would raise stability concerns. We remedy this in Section 4 through replacement rules for differential operators and arrive at an expansion, all terms of which are of the Jordan form $\mathrm{i}^{k+1} \langle f \rangle_k := \mathrm{i}^{k+1} \left(f \circ \partial_x^k + \partial_x^k \circ f \right)/2$, which makes them skew-Hermitian. These terms discretise as skew-Hermitian matrices in $\mathfrak{u}_M(\mathbb{C})$ whose exponentials are guaranteed to be unitary, whereby unitary evolution of the wavefunction and unconditional stability of the method are assured.

Keeping the eventual discretisation in mind, where $M = \mathcal{O}\left(\varepsilon^{-1}\right)$ degrees of spatial freedom are necessitated by the nature of the Schrödinger equation, we use the shorthand $\partial_x = \mathcal{O}\left(\varepsilon^{-1}\right)$ (although in principle it is an infinite-dimensional unbounded operator). We assume that the time step h scales as $h = \mathcal{O}\left(\varepsilon^\sigma\right)$ for some $0 < \sigma \leq 1$, allowing us to analyse all the terms solely in powers of ε. We find that, in principle, an effective scheme can be developed for any $\sigma > 1/3$. We are able to use time steps as large as $h = \mathcal{O}\left(\varepsilon^{1/2}\right)$, for instance, where we fall in the regime $h\|\mathcal{K}\| = \mathcal{O}\left(\varepsilon^{-1/2}\right)$, which represents a considerable improvement. In Section 5 we develop truncated Magnus expansions that can achieve an arbitrarily high order, expressed in powers of the semiclassical parameter ε.

We recap the Zassenhaus splitting algorithm in Section 6 which allows us to separate components of the Magnus expansion by powers of ε, arriving at asymptotic splittings of the form

$$u^{n+1} = \mathrm{e}^{\frac{1}{2}W^{[0]}} \mathrm{e}^{\frac{1}{2}W^{[1]}} \cdots \mathrm{e}^{\frac{1}{2}W^{[s]}} \mathrm{e}^{\mathcal{W}^{[s+1]}} \mathrm{e}^{\frac{1}{2}W^{[s]}} \cdots \mathrm{e}^{\frac{1}{2}W^{[1]}} \mathrm{e}^{\frac{1}{2}W^{[0]}} u^n,$$

which commit $\mathcal{O}\left(\varepsilon^{(s+3)\sigma-1}\right)$ error. Here $W^{[0]}, W^{[1]} = \mathcal{O}\left(\varepsilon^{\sigma-1}\right)$ and $W^{[2]} = \mathcal{O}\left(\varepsilon^{3\sigma-1}\right)$. The absence of an $\mathcal{O}\left(\varepsilon^{2\sigma-1}\right)$ term is an anomaly here since the following exponents

$W^{[k]}$, for $k \geq 3$, are $\mathcal{O}\left(\varepsilon^{(k+1)\sigma-1}\right)$. This is to be compared with the splittings of (Bader et al. 2014) which do not feature any exponent of size $\mathcal{O}\left(\varepsilon^{(2k)\sigma-1}\right)$.

In the approach presented here, evaluation of integrals is postponed till the very end, and terms of the splitting, $W^{[k]}$, still feature integrals at this stage. A range of options available to us for evaluation of integrals is considered in Section 6. If an analytic form of the interaction potential V is available, it might be possible to evaluate the integrals exactly. In general, it might be possible to approximate the integral through analytic means, standard quadratures or high frequency quadratures in special cases.

When dealing with highly oscillatory potentials, keeping the integrals intact helps suppress high-frequency oscillations. In contrast, approaches where the Magnus expansion commences with a Taylor expansion or a standard quadrature would require dramatic suppression of time-step. When the interaction potential V possesses more favourable temporal behaviour, values of V at only a few temporal grid points are required for a high order method once we resort to Gauss–Legendre quadrature or a Taylor expansion of $V(h)$. For $\mathcal{O}\left(\varepsilon^{7\sigma-1}\right)$ accuracy, for instance, we require merely three Gauss–Legendre knots, $t_k = h(1 + k\sqrt{3/5})/2, k = -1, 0, 1$.

Since we deliberately postpone spatial discretisation, the splittings are in an operatorial form, featuring undiscretised operators such as ∂_x^2. The choice of spatial discretisation at this stage does allow us some flexibility. However, we restrict our attention to spectral collocation, whereby ∂_x is replaced by a circulant matrix $\mathcal{K}$. The largest exponents in the splitting, $W^{[0]}$ and $W^{[1]}$, are of the form $\mathrm{i}h\partial_x^2$ and $\mathrm{i}h\mu(h)$, where $\mu(h)$ is an integral of the potential V from 0 to h. These are discretised as a circulant and a diagonal matrix, respectively, and are easily exponentiated with an $\mathcal{O}(M \log M)$ cost. The remaining exponents, which possess more complicated structures of the form $\mathrm{i}^{k+1} \langle f \rangle_k$, are small enough to be exponentiated effectively through Krylov methods such as Lanczos iterations (Tal Ezer & Kosloff 1984, Gallopoulos & Saad 1992) with an $\mathcal{O}(M \log M)$ cost per exponent. At this stage we have a fully concrete numerical scheme. We wrap up Section 6 by presenting numerical experiments to demonstrate the efficacy of our approach.

In Section 7 we exploit time symmetry of the Magnus expansion to derive splittings where the exponents $W^{[k]}$ are $\mathcal{O}\left(\varepsilon^{(2k-1)\sigma-1}\right)$ for $k \geq 2$. In these new splittings terms of size $\mathcal{O}\left(\varepsilon^{(2k)\sigma-1}\right)$ do not feature, reducing the number of exponents in the splitting (and the cost) to half and making higher order splittings simpler and more feasible. In order to achieve this feat we need to analyse the terms of the Magnus expansion in terms of their odd and even components. The time symmetry of the Magnus expansion results in the Magnus expansion being odd, whereby we are able to discard all its even components.

In an alternative approach, which is under parallel development, we commence our analysis from an integral-free approximation of the Magnus expansion. Here the integrals appearing in the Magnus expansion are replaced by Gauss–Legendre quadratures or Taylor expansions of V at the outset. The analysis in this approach, which will feature in another publication, should be easier to follow.

Since we wish to retain integrals till the very end in the approach presented here, the mathematical machinery we need to introduce becomes more involved. However, we end up with a highly flexible method – not only is it possible to approximate

the integrals through any quadrature method, but we may also use exact integrals for potentials which possess an analytic form. This flexibility allows us to tackle effectively potentials with weaker temporal regularity as well as highly oscillatory potentials of certain forms.

2 The Magnus expansion

For a general equation of the form (1.6) where $A(t)$ resides in a Lie algebra $\mathfrak{g}$, the central idea of the Magnus expansion (Magnus 1954) is to seek a solution for the flow (which must reside in the corresponding Lie group $\mathcal{G}$) as the exponential of an element $\Theta(t)$ in the Lie algebra $\mathfrak{g}$,

$$u(t) = \mathrm{e}^{\Theta(t)} u(0). \tag{2.1}$$

$\Theta(t)$ is expanded as an infinite series $\Theta(t) = \sum_{k=1}^{\infty} \Theta^{[k]}(t)$, the convergence of which is only guaranteed for sufficiently small time steps (Moan & Niesen 2008, Hochbruck & Lubich 2002). In practice, we work with finite truncations of this series. In order to keep truncation errors small and keeping the convergence criteria in mind, it is customary to evolve the solution in small time steps h,

$$u(t+h) = \mathrm{e}^{\Theta(t+h,t)} u(t), \tag{2.2}$$

starting from the initial step,

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

Here $\exp\left(\Theta(t+h,t)\right)$ is the operator which evolves the solution from t to $t+h$. Since it encodes the flow at time t under A, $\Theta(t+h,t)$ is recovered from $\Theta(h,0)$ by replacing all occurrences of $A(\zeta)$ with $A(t+\zeta)$. For all intents and purposes, therefore, it suffices to restrict the analysis to the first step (2.3). We hide the dependence on t for the larger part of this paper, shortening $\Theta(h,0)$ to $\Theta(h)$.

Simple differentiation of (2.3) together with elementary algebra (Iserles, Munthe-Kaas, Nørsett & Zanna 2000) show that the exponent has to satisfy the *dexpinv equation*,

$$\Theta'(h) = \mathrm{dexp}^{-1}_{\Theta(h)} A(h) = \sum_{m=0}^{\infty} \frac{B_m}{m!} \mathrm{ad}^m_{\Theta(h)} A(h), \tag{2.4}$$

where B_m are Bernoulli numbers and the adjoint map is recursively defined by $\mathrm{ad}^0_A(B) = B$, $\mathrm{ad}^{k+1}_A(B) = [A, \mathrm{ad}^k_A(B)]$. Magnus (1954) resorted to solving the dexpinv equation via Picard iteration,

$$\begin{aligned} \Omega_0(h) &= 0, \\ \Omega_{k+1}(h) &= \int_0^h \mathrm{dexp}^{-1}_{\Omega_k(\xi)} A(\xi) d\xi = \sum_{m=0}^{\infty} \frac{B_m}{m!} \int_0^h \mathrm{ad}^m_{\Omega_k(\xi)} A(\xi) d\xi, \end{aligned}$$

whereby $\Theta(h) = \lim_{k\to\infty} \Omega_k(h)$. The series $\Omega_k(h)$ is known as the Magnus expansion. For computational reasons, however, we adopt another version of this expansion described by Iserles & Nørsett (1999) where the operator $\Theta(h)$ is presented as an infinite

series: $\Theta(h) = \sum_{k=1}^{\infty} \Theta^{[k]}(h)$, where

$$\begin{aligned}
\Theta^{[1]}(h) =& \int_0^h A(\xi_1)d\xi_1,\\
\Theta^{[2]}(h) =& -\frac{1}{2}\int_0^h \left[\int_0^{\xi_1} A(\xi_2)d\xi_2, A(\xi_1)\right] d\xi_1,\\
\Theta^{[3]}(h) =& \frac{1}{12}\int_0^h \left[\int_0^{\xi_1} A(\xi_2)d\xi_2, \left[\int_0^{\xi_1} A(\xi_2)d\xi_2, A(\xi_1)\right]\right] d\xi_1\\
&+\frac{1}{4}\int_0^h \left[\int_0^{\xi_1}\left[\int_0^{\xi_2} A(\xi_3)d\xi_3, A(\xi_2)\right] d\xi_2, A(\xi_1)\right] d\xi_1,\\
\Theta^{[4]}(h) =& -\frac{1}{24}\int_0^h \left[\int_0^{\xi_1}\left[\int_0^{\xi_2} A(\xi_3)d\xi_3, \left[\int_0^{\xi_2} A(\xi_3)d\xi_3, A(\xi_2)\right]\right] d\xi_2, A(\xi_1)\right] d\xi_1\\
&-\frac{1}{24}\int_0^h \left[\int_0^{\xi_1}\left[\int_0^{\xi_2} A(\xi_3)d\xi_3, A(\xi_2)\right] d\xi_2, \left[\int_0^{\xi_1} A(\xi_2)d\xi_2, A(\xi_1)\right]\right] d\xi_1\\
&-\frac{1}{24}\int_0^h \left[\int_0^{\xi_1} A(\xi_2)d\xi_2, \left[\int_0^{\xi_1}\left[\int_0^{\xi_2} A(\xi_3)d\xi_3, A(\xi_2)\right] d\xi_2, A(\xi_1)\right]\right] d\xi_1\\
&-\frac{1}{8}\int_0^h \left[\int_0^{\xi_1} A(\xi_2)d\xi_2, \left[\int_0^{\xi_1} A(\xi_2)d\xi_2, \left[\int_0^{\xi_1} A(\xi_2)d\xi_2, A(\xi_1)\right]\right]\right] d\xi_1.
\end{aligned}$$

The terms in the Magnus expansion can be obtained through a recursive procedure, whereby they can be easily coded as binary rooted trees. Let $\mathcal{C}$ be a mapping from trees to terms. We recursively define the set of trees $\hat{\mathbb{T}}_k$ and the corresponding terms:

(1) Let $\hat{\mathbb{T}}_1 = \{\tau_0\}$, and $\mathcal{C}_{\tau_0}(h) = A(h)$.

(2) If $\tau_1 \in \hat{\mathbb{T}}_{m_1}$ and $\tau_2 \in \hat{\mathbb{T}}_{m_2}$ then there exists $\tau \in \hat{\mathbb{T}}_{m_1+m_2}$ such that

$$\mathcal{C}_\tau(h) = \left[\int_0^h \mathcal{C}_{\tau_1}(\xi)d\xi, \mathcal{C}_{\tau_2}(h)\right].$$

We depict the inverse of the bijection $\mathcal{C}$ as $\rightsquigarrow$, so that $\mathcal{C}_\tau \rightsquigarrow \tau$ for any tree τ. A graphical representation for these terms is obtained by representing the atomic expression $A(h)$ as a single vertex, $\tau_0 = \bullet$,

$$A(h) \rightsquigarrow \bullet\ ,$$

representing the unary operator of integration by a vertical line,

$$\int_0^h \mathcal{C}_\tau(\xi)\,\mathrm{d}\xi \rightsquigarrow \overset{\tau}{\bullet}\ ,$$

and the Lie bracket – a binary operator – by a fork,

$$[\mathcal{C}_{\tau_1}(h), \mathcal{C}_{\tau_2}(h)] \rightsquigarrow \overset{\tau_1}{\diagdown}\!\!\bullet\!\!\overset{\tau_2}{\diagup}\ .$$

This results in a more transparent representation of Magnus expansion and what is far more comfortable for investigation of the terms or for implementation. The first three sets of trees formed in this way, $\hat{\mathbb{T}}_1, \hat{\mathbb{T}}_2$ and $\hat{\mathbb{T}}_3$, are,

$$A(h) \rightsquigarrow \bullet \quad \in \hat{\mathbb{T}}_1,$$

$$\left[\int_0^h A(\xi_2)d\xi_2, A(h)\right] \rightsquigarrow \quad \in \hat{\mathbb{T}}_2,$$

$$\left[\int_0^h A(\xi_2)d\xi_2, \left[\int_0^h A(\xi_2)d\xi_2, A(h)\right]\right] \rightsquigarrow \quad \in \hat{\mathbb{T}}_3,$$

$$\left[\int_0^h \left[\int_0^{\xi_2} A(\xi_3)d\xi_3, A(\xi_2)\right] d\xi_2, A(h)\right] \rightsquigarrow \quad \in \hat{\mathbb{T}}_3.$$

It should be evident that trees in $\hat{\mathbb{T}}_k$ have $k-1$ vertical lines. Finally, we define the set of trees which actually appear in the Magnus expansion: $\mathbb{T}_k$,

$$\tau = \overset{\hat{\tau}}{\bullet} \in \mathbb{T}_k, \quad \mathcal{C}_\tau(h) = \int_0^h \mathcal{C}_{\hat{\tau}}(\xi)\,\mathrm{d}\xi, \quad \forall \hat{\tau} \in \hat{\mathbb{T}}_k,$$

so that every tree τ in $\mathbb{T}_k$ is obtained by adding an integral to a tree $\hat{\tau}$ from the auxiliary set $\hat{\mathbb{T}}_k$.

Each tree in $\mathbb{T}_k$ possesses k integrals. For $A(h) = \mathcal{O}\left(h^0\right)$ and $\tau \in \mathbb{T}_k$, it immediately follows that $\mathcal{C}_\tau(h) = \mathcal{O}\left(h^k\right)$. We say that the tree τ is $\mathcal{O}\left(h^k\right)$, for short. Similarly, every $\tau \in \hat{\mathbb{T}}_k$ is $\mathcal{O}\left(h^{k-1}\right)$. Each component of the Magnus expansion $\Theta^{[k]}(h)$ is composed solely of trees from the corresponding set $\mathbb{T}_k$,

$\Theta^{[1]}(h) =$,

$\Theta^{[2]}(h) = -\frac{1}{2}$,

$\Theta^{[3]}(h) = \frac{1}{12}$ $+ \frac{1}{4}$,

$\Theta^{[4]}(h) = -\frac{1}{24}$ $-\frac{1}{24}$ $-\frac{1}{24}$ $-\frac{1}{8}$.

The set of all trees that appear in the Magnus expansion is $\bigcup_{k\geq 1}\mathbb{T}_k$ and the Magnus expansion can be written in the form

$$\Theta(h)=\sum_{k=1}^{\infty}\Theta^{[k]}(h)=\sum_{k=1}^{\infty}\sum_{\tau\in\mathbb{T}_k}\alpha(\tau)\mathcal{C}_\tau(h),\ h\geq 0, \tag{2.5}$$

where $\alpha(\tau)$ is a scalar which can be recursively obtained (Iserles et al. 2000, Iserles & Nørsett 1999). The reader is forewarned about certain conflict of notation: the sets $\mathbb{T}_k$ in (Iserles et al. 2000, Iserles & Nørsett 1999) correspond to our auxiliary sets $\hat{\mathbb{T}}_{k+1}$ and the last integral occurs explicitly in the Magnus expansion corresponding to (2.5). A few discrepancies will, therefore, be found in numbering of trees and truncations of the Magnus expansion when directly comparing with these texts.

A tree τ is said to be of *power k in h* if k is the *greatest* integer such that $\mathcal{C}_\tau(h)=\mathcal{O}\left(h^k\right)$. We define $\mathbb{F}_k\subseteq\bigcup_{j\geq 1}\mathbb{T}_j$ as the set of all trees of power k in h which appear in the Magnus expansion. It is clear that $\tau\in\mathbb{T}_k$ implies that $\tau\in\mathbb{F}_m$ for some $m\geq k$ since a tree with k integrals is, at the very least, $\mathcal{O}\left(h^k\right)$. The two sets are not identical, however. For instance, it can be shown that the tree

belongs to $\mathbb{T}_2$ and $\mathbb{F}_3$. Such a gain in power occurs wherever we encounter a pattern of the form $\left[\int_0^h\mathcal{C}_\tau(\xi)\,\mathrm{d}\xi,\mathcal{C}_\tau(h)\right]$ with $\tau\in\hat{\mathbb{T}}_k$ which corresponds to the tree structure,

Upon expanding $\mathcal{C}_\tau(h)$ as $\sum_{i=k-1}^{\infty}h^i\mathrm{C}_i$ we find that the largest term in the above tree, $\left[\frac{1}{k}h^k\mathrm{C}_{k-1},h^{k-1}\mathrm{C}_{k-1}\right]$, vanishes since C_{k-1} commutes with itself (Iserles et al. 2000).

The *power truncated* Magnus expansion, which is based on truncation by the sets $\mathbb{F}_k$, is defined as

$$\Theta_p(h):=\sum_{k=1}^{p}\sum_{\tau\in\mathbb{F}_k}\alpha(\tau)\mathcal{C}_\tau(h). \tag{2.6}$$

The largest terms that have been discarded in this truncation correspond to trees from $\mathbb{F}_{p+1}$, which are $\mathcal{O}\left(h^{p+1}\right)$, so that this truncated expansion incurs an error of

$$\Theta_p(h)=\Theta(h)+\mathcal{O}\left(h^{p+1}\right),$$

where $\Theta(h)$ is the full Magnus series.

3 Expanding in an algebra of operators

The vector field in the Schrödinger equation (1.5) is a linear combination of the action of two operators, ∂_x^2 and the multiplication by the interaction potential $V(\xi)$, for any

$\xi \geq 0$. Since the Magnus expansion requires nested commutation, the focus of our interest is the free Lie algebra

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

i.e. the linear-space closure of all nested commutators generated by ∂_x^2 and $V(\cdot)$. To compute commutators we need in principle to describe their action on functions, e.g.

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

implies that $[V, \partial_x^2] = -(\partial_x^2 V) - 2(\partial_x V)\partial_x$, where we have suppressed dependence on the time variable ξ since it plays no role. In general, we note that all terms in $\mathfrak{F}$ belong to the set

$$\mathfrak{G} = \left\{ \sum_{k=0}^{n} f_k(x)\partial_x^k \,:\, n \in \mathbb{Z}_+,\ f_0, \ldots, f_n \in \mathrm{C}_p^\infty([-1,1];\mathbb{R}) \right\},$$

which is itself a Lie algebra (Bader et al. 2014) with the commutator

$$\begin{aligned}\left[\sum_{i=0}^{n} f_i(x)\partial_x^i, \sum_{j=0}^{m} g_j(x)\partial_x^j \right] = &\sum_{i=0}^{n}\sum_{j=0}^{m}\sum_{\ell=0}^{i} \binom{i}{\ell} f_i(x) \left(\partial_x^{i-\ell} g_j(x)\right) \partial_x^{\ell+j} \\ &- \sum_{j=0}^{m}\sum_{i=0}^{n}\sum_{\ell=0}^{j} \binom{j}{\ell} g_j(x) \left(\partial_x^{j-\ell} f_i(x)\right) \partial_x^{\ell+i}. \end{aligned} \tag{3.1}$$

Once we begin to simplify commutators of terms in the Lie algebra $\mathfrak{G}$ using the above simplification procedure, we encounter systematic reduction in differential operators which is summarised in the observation of *height reduction*. This observation is what justifies working directly in the undiscretised operators – it leads to a reduction in the spectral radius of terms, eventually allowing an effective asymptotic splitting.

Definition 1 *The height of a term in the free Lie algebra $\mathfrak{G}$ is defined as the degree of the highest-degree differential operator occurring in the term,*

$$\mathrm{ht}\left(\sum_{k=0}^{n} f_k(x)\partial_x^k \right) = n.$$

We note that the height of $(\partial_x^3 V)\partial_x + V\partial_x^2$, defined in this way, is two, not three, since ∂_x^3 is not acting as an operator but instead describes the third derivative of V. The term $0 \in \mathfrak{G}$ is assigned a height of -1, making it the only term with negative height.

We extend the notion of height to commutators in the free Lie algebra $\mathfrak{F}$ by defining it as the height of the term in $\mathfrak{G}$ to which the commutator reduces upon applying the simplification rule (3.1). Additionally, we may extend the notion of height to other algebras including the free algebra of ∂_x^2 and V under operatorial composition $\circ$ and their free Jordan algebra (which is the free algebra under the Jordan product, $A \bullet B := \frac{1}{2}(A \circ B + B \circ A)$).

Lemma 1 (Height reduction) *For all $C_1, C_2 \in \mathfrak{G}$, such that $C_1, C_2 \neq 0$,*

$$\mathrm{ht}([C_1, C_2]) \leq \mathrm{ht}(C_1) + \mathrm{ht}(C_2) - 1. \tag{3.2}$$

Proof Let $\mathrm{ht}(C_1) = n$ and $\mathrm{ht}(C_2) = m$, so that for some f_i and g_j, $C_1 = \sum_{i=0}^{n} f_i(x)\partial_x^i$ and $C_2 = \sum_{j=0}^{m} g_j(x)\partial_x^j$. The commutator $[C_1, C_2]$ is then described exactly by (3.1), where the terms featuring ∂_x^{n+m} in the two summations cancel out. The highest degree differential operator, therefore, has a degree not exceeding $n+m-1$. □

We note that in general the terms corresponding to ∂_x^{n+m-1} do not cancel out and, with the exception of some special cases, (3.2) holds as an equality.

This property extends in a natural way to nested commutators appearing in the Magnus expansion. Terms in the Magnus expansion (2.5) are of the form

$$I_{\mathcal{S},\boldsymbol{L}}(h) = \int_{\mathcal{S}} \boldsymbol{L}(\xi_1, \xi_2, \ldots, \xi_s)\,\mathrm{d}\boldsymbol{\xi},$$

where $\boldsymbol{L}$ a multilinear form free of integrals and consisting solely of commutators featuring $A(\xi_j)$, $j = 1, 2, \ldots, s$, while $\mathcal{S}$ is an s-dimensional polytope of a special form,

$$\mathcal{S} = \{\boldsymbol{\xi} \in \mathbb{R}^s \ : \ \xi_1 \in [0, h], \quad \xi_l \in [0, \xi_{m_l}], \quad l = 2, 3, \ldots, s\}, \tag{3.3}$$

where $m_l \in \{1, 2, \ldots, l-1\}$, $l = 2, 3, \ldots, s$. For instance, the multilinear form occurring in the tree

is $\boldsymbol{L} = [A(\xi), A(\zeta)]$ and the polytope of integration is the triangle $\mathcal{S} = \{(\zeta, \xi) \in \mathbb{R}^2 \ : \ \zeta \in [0, h], \quad \xi \in [0, \zeta]\}$. Simplification identities of the free Lie algebra such as (3.1), which solely concern themselves with the algebraic structure in the spatial domain, can be applied directly to simplify $\boldsymbol{L}$.

Corollary 1 *For any $C_1, \ldots, C_n \in \{\mathrm{i}\partial_x^2\} \cup \{\mathrm{i}V(\xi) \ : \ \xi \in \mathbb{R}\}$ – where we take the imaginary unit into account – if $\boldsymbol{L}(C_1, \ldots, C_n) \neq 0$,*

$$\mathrm{ht}\left(\boldsymbol{L}(C_1, \ldots, C_n)\right) \leq \sum_{k=1}^{n} \mathrm{ht}(C_k) - (n-1),$$

where $\boldsymbol{L}$ is a commutator involving $C_1, \ldots, C_n$ in any order.

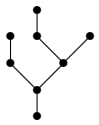
Where negative height is encountered in a subpart of the commutator $\boldsymbol{L}$, the entire term vanishes. Just like the case of Lemma 1, there are choices of V such that Corollary 1 holds with equality.

Commutators in the Magnus expansion $\Theta(h) = \sum_{k=1}^{\infty} \Theta^{[k]}(h)$ can be expanded in $\mathfrak{G}$. With $A(\zeta) = \mathrm{i}\varepsilon\partial_x^2 + \mathrm{i}\varepsilon^{-1}V(\zeta)$, the first term of the Magnus expansion is the integral

 : $$\int_0^h A(\zeta)\,\mathrm{d}\zeta = \int_0^h \left(\mathrm{i}\varepsilon\partial_x^2 + \mathrm{i}\varepsilon^{-1}V(\zeta)\right)\,\mathrm{d}\zeta = \mathrm{i}h\varepsilon\partial_x^2 + \mathrm{i}\varepsilon^{-1}\int_0^h V(\zeta)\,\mathrm{d}\zeta. \tag{3.4}$$

Other terms can be written as nested integrals of commutators $\boldsymbol{L}(A_1,\dots,A_n)$, $A_j = A(\xi_j) = \mathrm{i}\varepsilon\partial_x^2 + \mathrm{i}\varepsilon^{-1}V(\xi_j)$, which can again be worked out in the free Lie algebra,

$$
\begin{aligned}
&: \int_0^h \left[\int_0^\zeta A(\xi)\,\mathrm{d}\xi\ ,\ A(\zeta)\right]\,\mathrm{d}\zeta = \int_0^h\int_0^\zeta [A(\xi),A(\zeta)]\,\mathrm{d}\xi\,\mathrm{d}\zeta\\
&\quad= \int_0^h\int_0^\zeta \left[\mathrm{i}\varepsilon\partial_x^2 + \mathrm{i}\varepsilon^{-1}V(\xi)\ ,\ \mathrm{i}\varepsilon\partial_x^2 + \mathrm{i}\varepsilon^{-1}V(\zeta)\right]\,\mathrm{d}\xi\,\mathrm{d}\zeta\\
&\quad= -\int_0^h\int_0^\zeta \left[\partial_x^2, V(\zeta)\right] + \left[V(\xi),\partial_x^2\right]\,\mathrm{d}\xi\,\mathrm{d}\zeta\\
&\quad= 2\left(\int_0^h\int_0^\zeta (\partial_x V(\xi))\,\mathrm{d}\xi\,\mathrm{d}\zeta - \int_0^h \zeta(\partial_x V(\zeta))\,\mathrm{d}\zeta\right)\partial_x\\
&\qquad+ \int_0^h\int_0^\zeta (\partial_x^2 V(\xi))\,\mathrm{d}\xi\,\mathrm{d}\zeta - \int_0^h \zeta(\partial_x^2 V(\zeta))\,\mathrm{d}\zeta,
\end{aligned}
\tag{3.5}
$$



$$
\begin{aligned}
&: 4\mathrm{i}\varepsilon\left(\int_0^h \zeta\int_0^\zeta (\partial_x^2 V(\xi))\,\mathrm{d}\xi\,\mathrm{d}\zeta - \int_0^h \zeta^2(\partial_x^2 V(\zeta))\,\mathrm{d}\zeta\right)\partial_x^2\\
&\quad+ 4\mathrm{i}\varepsilon\left(\int_0^h \zeta\int_0^\zeta (\partial_x^3 V(\xi))\,\mathrm{d}\xi\,\mathrm{d}\zeta - \int_0^h \zeta^2(\partial_x^3 V(\zeta))\,\mathrm{d}\zeta\right)\partial_x\\
&\quad+ 2\mathrm{i}\varepsilon^{-1}\left(\int_0^h \zeta(\partial_x V(\zeta))\int_0^\zeta (\partial_x V(\xi))\,\mathrm{d}\xi\,\mathrm{d}\zeta - \int_0^h \left(\int_0^\zeta (\partial_x V(\xi))\,\mathrm{d}\xi\right)^2\,\mathrm{d}\zeta\right)\\
&\quad+ \mathrm{i}\varepsilon\left(\int_0^h \zeta\int_0^\zeta (\partial_x^4 V(\xi))\,\mathrm{d}\xi\,\mathrm{d}\zeta - \int_0^h \zeta^2(\partial_x^4 V(\zeta))\,\mathrm{d}\zeta\right).
\end{aligned}
\tag{3.6}
$$

After simplifying terms in the Magnus expansion (2.5) we arrive at expressions such as (3.5) and (3.6), where each integral is of the form

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

where $\boldsymbol{P}(\boldsymbol{\xi}) = \prod_{j=1}^s P_j(\xi_j)$ for some P_j, and $\mathcal{S}$ is an s-dimensional polytope of the special form (3.3).

Integration over the polytope $\mathcal{S}$ in the temporal domain has remained an afterthought up to this stage. The special forms of these polytopes, and of certain 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 iden-

tities:

$$\int_0^h P_1(\xi_1)\left(\int_0^{\xi_1} P_2(\xi_2)d\xi_2\right)d\xi_1 = \int_0^h P_2(\xi_1)\left(\int_{\xi_1}^h P_1(\xi_2)d\xi_2\right)d\xi_1, \tag{3.7}$$

$$\int_0^h P_1(\xi_1)d\xi_1\left(\int_0^{\xi_1} P_2(\xi_2)d\xi_2\right)\left(\int_0^{\xi_1} P_3(\xi_3)d\xi_3\right)d\xi_1 \tag{3.8}$$

$$= \int_0^h \left(\int_{\xi_3}^h P_1(\xi_1)d\xi_1\right)\left(P_2(\xi_3)\int_0^{\xi_3} P_3(\xi_2)d\xi_2 + P_3(\xi_3)\int_0^{\xi_3} P_2(\xi_2)d\xi_2\right)d\xi_3.$$

In our simplifications, (3.5) and (3.6), 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 $P_1(\xi_1)=1$, $P_2(\xi_2)=\partial_x V(\xi_2)$ and $P_1(\xi_1)=\xi_1$, $P_2(\xi_2)=\partial_x V(\xi_2)$, 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 $P_1(\xi_1)=1$, $P_2(\xi_2)=\partial_x V(\xi_2)$, $P_3(\xi_3)=\partial_x V(\xi_3)$.

Although it might be possible to develop a 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 case should also be relatively straightforward.

4 Regaining stability

The algebraic structure of the Lie algebra $\mathfrak{G}$ is far too weak for our stability concerns. Expanding nested commutators in the Lie algebra $\mathfrak{G}$ does give us a commutator-free method, but it does so by mixing all powers of ∂_x: in the simplification of the first three trees – (3.4), (3.5) and (3.6) – we encounter terms of the form $\mathrm{i}\partial_x^2, \mathrm{i}f(x), f(x), f(x)\partial_x$ and $\mathrm{i}f(x)\partial_x^2$ for some real-valued function $f\in \mathrm{C}_p^\infty([-1,1];\mathbb{R})$. This presence is worrisome for an important reason, namely *stability.*

Both ∂_x^2 and multiplication by $V(x,t)$ are Hermitian operators, therefore $A(t)=\mathrm{i}\varepsilon\partial_x^2+\mathrm{i}\varepsilon^{-1}V(x,t)$ is a skew-Hermitian operator: its flow is thus unitary. This survives under eventual discretisation, because any reasonable approximation of ∂_x^2 under periodic boundaries preserves Hermitian structure. However, terms of the form $f(x)$, without the leading i are Hermitian while terms of the form $f(x)\partial_x$ and $\mathrm{i}f(x)\partial_x^2$, are in general neither skew-Hermitian nor Hermitian. This is fraught with loss of unitarity and stability.

In similar vein to Bader et al. (2014), in the pursuit of stability, we proceed to

replace all odd powers of ∂_x that are accompanied by i using the identities

$$
\begin{aligned}
f\partial_x &= -\tfrac{1}{2}\left[\int_0^x f(\xi)\,\mathrm{d}\xi\right]\partial_x^2 - \tfrac{1}{2}\partial_x f + \tfrac{1}{2}\partial_x^2\left[\int_0^x f(\xi)\,\mathrm{d}\xi\ \cdot\right],\\
f\partial_x^3 &= -(\partial_x f)\partial_x^2 - \tfrac{1}{4}\left[\int_0^x f(\xi)\,\mathrm{d}\xi\right]\partial_x^4 + \tfrac{1}{4}\partial_x^3 f - \tfrac{1}{2}\partial_x^2[(\partial_x f)\cdot] + \tfrac{1}{4}\partial_x^4\left[\int_0^x f(\xi)\,\mathrm{d}\xi\ \cdot\right],\\
f\partial_x^5 &= \tfrac{4}{3}(\partial_x^3 f)\partial_x^2 - \tfrac{5}{3}(\partial_x f)\partial_x^4 - \tfrac{1}{6}\left[\int_0^x f(\xi)\,\mathrm{d}\xi\right]\partial_x^6 - \tfrac{1}{2}\partial_x^5 f + \tfrac{7}{6}\partial_x^2[(\partial_x^3 f)\cdot]\\
&\quad - \tfrac{5}{6}\partial_x^4[(\partial_x f)\cdot] + \tfrac{1}{6}\partial_x^6\left[\int_0^x f(\xi)\,\mathrm{d}\xi\ \cdot\right],
\end{aligned}
$$

where f is any C^1 function. The general form for expressing $f\partial_x^{2s+1}$ as a linear combination of even derivatives is also known (Bader et al. 2014).

In the Zassenhaus splitting for time-independent potentials (Bader et al. 2014), the commutators arise solely from the *symmetric Baker–Campbell–Hausdorff formula* where each commutator has an odd number of *letters*. In the case of the Schrödinger equation, where the operators ∂_x^2 and V are accompanied by i, this translates into an odd power of i for each commutator.

The Magnus expansion, however, does not posses such a desirable structure – it has commutators with odd as well as even number of letters. As a consequence, we have odd and even powers of i accompanying our terms and it does not suffice to automatically replace odd powers of ∂_x. Instead, we replace all odd powers of ∂_x when accompanied by an odd power of i and all even powers of ∂_x when accompanied by an even power of i. A general formula for replacement of even derivatives by odd derivatives can be proven along similar lines to (Bader et al. 2014). For all practical purposes, however, we only require the identities,

$$
\begin{aligned}
f &= -\left[\int_0^x f(\xi)\,\mathrm{d}\xi\right]\partial_x + \partial_x\left[\int_0^x f(\xi)\,\mathrm{d}\xi\,\cdot\right],\\
f\partial_x^2 &= -\tfrac{1}{3}\left[\int_0^x f(\xi)\,\mathrm{d}\xi\right]\partial_x^3 - \tfrac{2}{3}(\partial_x f)\partial_x - \tfrac{1}{3}\partial_x[(\partial_x f)\cdot] + \tfrac{1}{3}\partial_x^3\left[\int_0^x f(\xi)\,\mathrm{d}\xi\,\cdot\right],\\
f\partial_x^4 &= -\tfrac{1}{5}\left[\int_0^x f(\xi)\,\mathrm{d}\xi\right]\partial_x^5 - \tfrac{4}{3}(\partial_x f)\partial_x^3 + \tfrac{8}{15}(\partial_x^3 f)\partial_x + \tfrac{7}{15}\partial_x[(\partial_x^3 f)\cdot]\\
&\quad - \tfrac{2}{3}\partial_x^3[(\partial_x f)\cdot] + \tfrac{1}{5}\partial_x^5\left[\int_0^x f(\xi)\,\mathrm{d}\xi\,\cdot\right],
\end{aligned}
$$

which can be easily verified. Since these replacement rules are identities and our definition of height requires simplification of all terms to terms residing in $\mathfrak{G}$, the heights of both sides of each identity are the same. Consequently, the property of height reduction from Lemma 1 and Corollary 1 is retained.

The motivation behind the observation of height reduction, however, is to study a systematic decrease in the highest degree differential operator occurring in a term. Upon eventual spatial discretisation the differential operators occurring in each expression are replaced by matrices, the spectral radius of which is largely governed by the highest degree differential operator occurring in it. The notion of height is supposed to provide a direct estimate of the spectral radius in this way. However,

the highest degree differential operator is a property of an expression and clearly the replacement rules seem to increase this by one.

However, we note in Lemma 2 that the correct use of these replacement rules does not increase the highest degree differential operator in the expressions that concern us. Since we work in the free Lie algebra generated by $\mathrm{i}\partial_x^2$ and $\mathrm{i}V$, the terms that interest us are commutators of the form $\boldsymbol{L}$ detailed in Corollary 1. We remind the reader that we only prescribe replacing odd-degree differential operators when accompanied by i and replacing even-degree differential operators when not accompanied by i.

Lemma 2 (Reduction of highest degree differential operators) *For any $C_j \in \{\mathrm{i}\partial_x^2\} \cup \{\mathrm{i}V(\xi) \ : \ \xi \in \mathbb{R}\}, j = 1, \ldots, n$ consider a commutator $\boldsymbol{L}(C_1, \ldots, C_n) \neq 0$ featuring C_j in any order. If the highest-degree differential operator occurring in the simplification of $\boldsymbol{L}$ after replacement of appropriate derivatives is ∂_x^d, then*

$$d \leq \sum_{k=1}^{n} \mathrm{ht}(C_k) - (n-1).$$

Proof We assume j instances of $\mathrm{i}\partial_x^2$ and $n-j$ instances of $\mathrm{i}V(\cdot)$ in the commutator $\boldsymbol{L}$. Height reduction implies $\mathrm{ht}(\boldsymbol{L}) \leq 2j+1-n$, since $\boldsymbol{L} \neq 0$. We argue that the derivative replacement rules of section 4, when correctly used, cannot produce terms with differential operators of a degree higher than $2j+1-n$.

Since the replacement rules of Section 4 never increase the degree of differential operators by more than one (Bader et al. 2014), we need only concern ourselves with the largest possible term, $\mathrm{i}^n f \partial_x^{2j+1-n}$ for some $f \in \mathrm{C}_p^\infty([-1,1];\mathbb{R})$. However, the term is either of the form $g\partial_x^{2m+1}$ or $\mathrm{i}g\partial_x^{2m}$ for some $m \in \mathbb{N}$ and $g \in \mathrm{C}_p^\infty([-1,1];\mathbb{R})$. In either case it does not warrant replacement. □

The consequence of Lemma 2 is that we continue to witness the same decrease in the highest degree differential operator despite the replacement rules, and the notion of height remains a valid proxy. We note that there are choices of V for which $\mathrm{ht}(\boldsymbol{L}) = 2j+1-n$ and Lemma 2 holds with equality.

Once the appropriate odd and even differential operators are replaced, operators of the form $f\partial_x^k + \partial_x^k[f\,\cdot\,]$ start appearing ubiquitously in our analysis. Far from being unique to the Magnus expansion, this is characteristic of the free Lie algebra of ∂_x^2 and V – these algebraic forms also appear in Zassenhaus splittings for time-independent potentials (Bader et al. 2014). We introduce a useful notation

$$\langle f \rangle_k := f \bullet \ \partial_x^k = \tfrac{1}{2}\left\{f \circ \partial_x^k + \partial_x^k \circ f\right\} = \tfrac{1}{2}\left\{f\partial_x^k + \partial_x^k[f\,\cdot\,]\right\}, \qquad f \in \mathrm{C}_p^\infty([-1,1];\mathbb{R}),$$

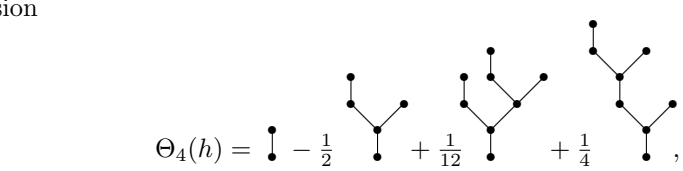
where $\bullet$ is the *Jordan product* on the associative algebra of $\circ$ (operatorial composition). The notion of height extends to such terms since it is possible to reduce them to expressions in $\mathfrak{G}$. A simplification procedure corresponding to (3.1) shows that,

$$\begin{aligned} \mathrm{ht}\left(\langle f \rangle_k\right) &= k, \\ \mathrm{ht}\left(\left[\langle f \rangle_k, \langle g \rangle_l\right]\right) &\leq k+l-1, \end{aligned}$$

for all $f, g \neq 0$ and $k, l \in \mathbb{N}_0$. It should be obvious that $\langle f \rangle_k$ is Hermitian if k is even and skew-Hermitian if k is odd. In this notation $\langle 1 \rangle_2 = \partial_x^2$ and $\langle V \rangle_0 = V$. It is worth

noting that there is rich algebraic theory behind these structures which is currently under development and will feature in another publication, but not much is lost here by considering these as merely a notational convenience.

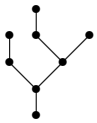
After suitable replacement of derivatives and applications of the integration identities (3.7) and (3.8), the trees in $\mathbb{F}_1, \mathbb{F}_3$ and $\mathbb{F}_4$ which appear in the truncated Magnus expansion

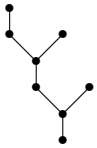
$\Theta_4(h) =$  (4.1)

simplify to the forms

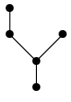
 :
$$\mathrm{i}h\varepsilon\partial_x^2 + \mathrm{i}\varepsilon^{-1}\int_0^h V(\zeta)\,\mathrm{d}\zeta, \tag{4.2}$$

 :
$$-4\left\langle \int_0^h \left(\zeta - \tfrac{h}{2}\right)\left(\partial_x V(\zeta)\right)\mathrm{d}\zeta \right\rangle_1, \tag{4.3}$$

 :
$$-2\mathrm{i}\varepsilon^{-1}\int_0^h\int_0^\zeta (2h-3\zeta)\left(\partial_x V(\zeta)\right)\left(\partial_x V(\xi)\right)\mathrm{d}\xi\,\mathrm{d}\zeta + 2\mathrm{i}\varepsilon\left\langle \int_0^h \left(h^2-3\zeta^2\right)\left(\partial_x^2 V(\zeta)\right)\mathrm{d}\zeta \right\rangle_2, \tag{4.4}$$

 :
$$2\mathrm{i}\varepsilon^{-1}\int_0^h\int_0^\zeta (\zeta-2\xi)\left(\partial_x V(\zeta)\right)\left(\partial_x V(\xi)\right)\mathrm{d}\xi\,\mathrm{d}\zeta - 2\mathrm{i}\varepsilon\left\langle \int_0^h \left(h^2-4h\zeta+3\zeta^2\right)\left(\partial_x^2 V(\zeta)\right)\mathrm{d}\zeta \right\rangle_2. \tag{4.5}$$

The term $\left\langle \int_0^h \left(\zeta - \frac{h}{2}\right)\left(\partial_x V(\zeta)\right)\mathrm{d}\zeta \right\rangle_1$, which occurs in the simplification of the tree

 $\in \mathbb{F}_3$,

might seem to be $\mathcal{O}\left(h^2\right)$ at first sight, contradicting our expectations from a tree in $\mathbb{F}_3$. A closer look at the special form of the integrand, however, shows that the term is indeed $\mathcal{O}\left(h^3\right)$. To observe this, consider $V(\zeta)$ expanded about $h = 0$ so that $V(h) = V(0) + \sum_{k=1}^{\infty} h^k V^{(k)}(0)/k!$ and note that the h^2 term $\int_0^h \left(\zeta - \frac{h}{2}\right)\left(\partial_x V(0)\right)\mathrm{d}\zeta$ vanishes. Similar care has to be exercised throughout the simplifications in combining the appropriate terms before analysing size.

Upon eventual discretisation through spectral methods, we replace the operator ∂_x by the skew-symmetric circulant matrix $\mathcal{K}$ whose action can be effectively computed through two FFTs. In the second tree we encounter an expression of the form $\langle f\rangle_1$ which is eventually discretised in the form $\frac{1}{2}\left(\mathcal{K}\mathcal{D}_f+\mathcal{D}_f\mathcal{K}\right)$, where $\mathcal{D}_f$ is a real diagonal matrix discretising $f(x)$. It, therefore, retains skew-Hermiticity.

In fact, our entire Magnus expansion is of the form $\sum_{k=0}^{\infty}\mathrm{i}^{k+1}\langle f_k\rangle_k$ for appropriate terms f_k, and it can be seen that each term $\mathrm{i}^{k+1}\langle f_k\rangle_k$ is skew-Hermitian. It is discretised in the skew-Hermitian form $\mathrm{i}^{k+1}\left(\mathcal{K}^k\mathcal{D}_{f_k}+\mathcal{D}_{f_k}\mathcal{K}^k\right)/2$. The Magnus expansion, developed in this way preserves skew-Hermiticity and its exponential preserves unitarity, which is desirable in the case of the Schrödinger equation but also gives us unconditional stability for free.

A *Wentzel–Kramers–Brillouin (WKB)* analysis (Hinch 1991) shows that the solution of the Schrödinger equation develops spatial oscillations of order $\mathcal{O}\left(\varepsilon^{-1}\right)$ (Jin et al. 2011, Bao, Jin & P. A. Markowich 2002). A reasonable approximation of the solution therefore necessitates taking $\mathcal{O}\left(\varepsilon^{-1}\right)$ Fourier modes and consequently ∂_x^n scales as $\mathcal{O}\left(\varepsilon^{-n}\right)$ upon discretisation. We analyse all terms in the common *currency* of the inherent semiclassical parameter ε and additionally assume that our choice of the time-step, h, is governed by $h=\mathcal{O}\left(\varepsilon^{\sigma}\right)$, for some $0<\sigma\leq 1$.

The height of a term, which serves as a proxy for the degree of the highest degree differential operator occurring in it, helps us estimate the spectral radius. For instance, the height of $\langle f\rangle_n$ is n and it scales like $\mathcal{O}\left(\varepsilon^{-n}\right)$ for $f=\mathcal{O}\left(\varepsilon^{0}\right)$. The property of height reduction can therefore be seen to result in a systematic reduction of spectral radius, highlighting the advantage of working with undiscretised operators.

5 Expanding in powers of the small parameter

The *power truncated* Magnus expansion (2.6),

$$\Theta_p(h):=\sum_{k=1}^{p}\sum_{\tau\in\mathbb{F}_k}\alpha(\tau)\mathcal{C}_\tau(h),$$

has the desirable characteristic of being odd in h due to the time symmetry of its flow (Iserles & Nørsett 1999, Iserles et al. 2000, Iserles, Nørsett & Rasmussen 2001). Odd-indexed methods of this form consequently gain an extra power of h,

$$\Theta_{2p-1}(h)=\Theta(h)+\mathcal{O}\left(h^{2p+1}\right). \tag{5.1}$$

Once we start analysing the Magnus expansion for the Schrödinger equation in the currency of ε we need to consider truncating the sets of trees by powers of ε. For this purpose we define the set $\mathbb{E}_k$ along (somewhat) similar lines to the definition of $\mathbb{F}_k$: $\tau\in\mathbb{E}_k$ if k is the *greatest* integer such that $\tau=\mathcal{O}\left(\varepsilon^{k\sigma-1}\right)$. It turns out that the sets $\mathbb{E}_k$ and $\mathbb{F}_k$ are, in fact, identical.

Theorem 3 $\mathbb{E}_m=\mathbb{F}_m$

Proof Consider any $\tau\in\mathbb{F}_m$. Clearly $\tau\in\mathbb{T}_n$ for some $n\leq m$, and we know that $\mathcal{C}_\tau(h)$ is a nested integral of a commutator $\tilde{\boldsymbol{L}}(A_1,\ldots,A_n)$, where $A_k=A(\xi_k)=$

$\mathrm{i}\varepsilon\partial_x^2 + \mathrm{i}\varepsilon^{-1}V(\xi_k)$. By linearity of the Lie bracket, this commutator expands to a linear combination of commutators of the form $\boldsymbol{L}(C_1, \ldots, C_n)$, where each $C_k \in \{\mathrm{i}\partial_x^2\} \cup \{\mathrm{i}V(\xi) \,:\, \xi \in \mathbb{R}\}$. Not all of these commutators end up vanishing, and we restrict our attention to those with non-negative height, $\boldsymbol{L}(C_1, \ldots, C_n) \neq 0$.

We assume j instances of $\mathrm{i}\partial_x^2$ and $n-j$ instances of $\mathrm{i}V(\cdot)$ in $\boldsymbol{L}$. From Corollary 1 and Lemma 2 we know that there is some V, $\mathcal{O}\left(h^0\right)$ and $\mathcal{O}\left(\varepsilon^0\right)$ in size, for which expansion of $\boldsymbol{L}$ in the Lie algebra (taking derivative replacement rules into account) results in the highest degree of a differential operator being $2j-(n-1)$.

Such a term is $\mathcal{O}\left(h^m\right)$ by definition of $\mathbb{F}_m$ and is accompanied by a scalar factor of ε^{2j-n}. Combining these facts, we see that the tree τ is $\mathcal{O}\left(\varepsilon^{m\sigma-1}\right)$ in size and consequently also belongs to $\mathbb{E}_m$.

Every $\tau \in \mathbb{E}_m$, of course, must be in some $\mathbb{F}_n$ and thus in $\mathbb{E}_n$. However, the definition of $\mathbb{E}_k$ dictates that $n = m$. Thus the sets $\mathbb{F}_m$ and $\mathbb{E}_m$ must coincide. □

The consequence of Theorem 3 is that Magnus expansions truncated by $\mathbb{E}_k$ (which interest us when analysing in powers of ε),

$$\Theta_p^\varepsilon(h) := \sum_{k=1}^{p} \sum_{\tau \in \mathbb{E}_k} \alpha(\tau) \mathcal{C}_\tau^\varepsilon(h), \tag{5.2}$$

are identical to the corresponding Magnus expansions (2.6) truncated by $\mathbb{F}_k$ (analysed in powers of h), and we may use Θ_p to denote both. It must be noted that under scalings which differ from our choice of $M = \mathcal{O}\left(\varepsilon^{-1}\right)$, this observation no longer holds. Here the superscript ε explicitly acknowledges the dependence on ε.

The largest trees that are discarded in this expansion are $\mathcal{O}\left(\varepsilon^{(p+1)\sigma-1}\right)$ since they belong to $\mathbb{E}_{p+1}$, and the expansion incurs an error of

$$\Theta_p^\varepsilon(h) = \Theta(h) + \mathcal{O}\left(\varepsilon^{(p+1)\sigma-1}\right). \tag{5.3}$$

Odd-indexed truncations, as expected, gain a power of ε^σ,

$$\Theta_{2p-1}^\varepsilon(h) = \Theta(h) + \mathcal{O}\left(\varepsilon^{(2p+1)\sigma-1}\right). \tag{5.4}$$

We must make a clear distinction between discarding *trees* by powers of ε and discarding *terms* obtained upon simplification of these trees in the free Lie algebra. A tree $\tau \in \mathbb{E}_m$ is $\mathcal{O}\left(\varepsilon^{m\sigma-1}\right)$ by definition, and is included in the expansion $\Theta_{2p-1}^\varepsilon$ if $m \leq 2p-1$. Upon simplifying, however, it might feature $\mathcal{O}\left(\varepsilon^{2p\sigma-1}\right)$, $\mathcal{O}\left(\varepsilon^{(2p+1)\sigma-1}\right)$ or smaller terms which must, nevertheless, be retained in the expansion $\Theta_{2p-1}^\varepsilon$ for the sake of time symmetry.

Once the desirable time symmetry properties are fully exploited, though, we discard all $\mathcal{O}\left(\varepsilon^{(2p+1)\sigma-1}\right)$ and smaller terms from $\Theta_{2p-1}^\varepsilon$ and arrive at $\tilde{\Theta}_{2p-1}^\varepsilon$, which carries an error of

$$\tilde{\Theta}_{2p-1}^\varepsilon(h) = \Theta_{2p-1}^\varepsilon(h) + \mathcal{O}\left(\varepsilon^{(2p+1)\sigma-1}\right) = \Theta(h) + \mathcal{O}\left(\varepsilon^{(2p+1)\sigma-1}\right). \tag{5.5}$$

We are prohibited, however, to discard any $\mathcal{O}\left(\varepsilon^{2p\sigma-1}\right)$ terms that might originate in simplifications without compromising on error. For completeness, we also define $\tilde{\Theta}_{2p}^\varepsilon$

as the expansion obtained after discarding all $\mathcal{O}\left(\varepsilon^{(2p+1)\sigma-1}\right)$ and smaller terms from $\Theta^{\varepsilon}_{2p}$. However, it is preferable to work solely with odd-indexed expansions due to the free gain in power. For instance, the Magnus expansion $\tilde{\Theta}^{\varepsilon}_4$, obtained by discarding all $\mathcal{O}\left(\varepsilon^{5\sigma-1}\right)$ terms from Θ_4 (4.1–5),

$$\begin{aligned}\tilde{\Theta}^{\varepsilon}_4(h) = \overbrace{\mathrm{i}h\varepsilon\partial_x^2 + \mathrm{i}\varepsilon^{-1}\int_0^h V(\zeta)\,\mathrm{d}\zeta}^{\mathcal{O}\left(\varepsilon^{\sigma-1}\right)} + \overbrace{2\left\langle\int_0^h \left(\zeta-\tfrac{h}{2}\right)\left(\partial_x V(\zeta)\right)\mathrm{d}\zeta\right\rangle_1}^{\mathcal{O}\left(\varepsilon^{3\sigma-1}\right)}\\ \overbrace{+\,\mathrm{i}\varepsilon^{-1}\int_0^h\int_0^\zeta \left(\zeta-\xi-\tfrac{h}{3}\right)\left[\partial_x V(\zeta)\right]\left[\partial_x V(\xi)\right]\,\mathrm{d}\xi\,\mathrm{d}\zeta}^{\mathcal{O}\left(\varepsilon^{4\sigma-1}\right)}\\ \overbrace{-\,2\mathrm{i}\varepsilon\left\langle\int_0^h\left(\zeta^2-h\zeta+\tfrac{h^2}{6}\right)\left(\partial_x^2 V(\zeta)\right)\mathrm{d}\zeta\right\rangle_2}^{\mathcal{O}\left(\varepsilon^{4\sigma-1}\right)},\end{aligned} \tag{5.6}$$

shares the same $\mathcal{O}\left(\varepsilon^{5\sigma-1}\right)$ error with the simpler expansion

$$\tilde{\Theta}^{\varepsilon}_3(h) = \overbrace{\mathrm{i}h\varepsilon\partial_x^2 + \mathrm{i}\varepsilon^{-1}\int_0^h V(\zeta)\,\mathrm{d}\zeta}^{\mathcal{O}\left(\varepsilon^{\sigma-1}\right)} + \overbrace{2\left\langle\int_0^h \left(\zeta-\tfrac{h}{2}\right)\left(\partial_x V(\zeta)\right)\mathrm{d}\zeta\right\rangle_1}^{\mathcal{O}\left(\varepsilon^{3\sigma-1}\right)}. \tag{5.7}$$

Before rushing on to make a conjecture that all terms of the form $\mathcal{O}\left(\varepsilon^{2k\sigma-1}\right)$, such as the $\mathcal{O}\left(\varepsilon^{4\sigma-1}\right)$ term in $\tilde{\Theta}^{\varepsilon}_4$, may be discarded at no cost in general, it is worth remembering that the increased order (5.4) due to time symmetry of an odd-indexed method, $\tilde{\Theta}^{\varepsilon}_{2p-1}$, only allows us to discard the 'penultimate' $\mathcal{O}\left(\varepsilon^{2p\sigma-1}\right)$ trees. Thus $\tilde{\Theta}^{\varepsilon}_5$ will once again feature the discarded $\mathcal{O}\left(\varepsilon^{4\sigma-1}\right)$ trees while being free to discard the $\mathcal{O}\left(\varepsilon^{6\sigma-1}\right)$ trees, and $\tilde{\Theta}^{\varepsilon}_7$ will feature trees of sizes $\mathcal{O}\left(\varepsilon^{k\sigma-1}\right)$ for $k = 1,3,4,5,6,7$ while being free to discard the $\mathcal{O}\left(\varepsilon^{8\sigma-1}\right)$ trees. Terms (to be contrasted with trees) of size $\mathcal{O}\left(\varepsilon^{8\sigma-1}\right)$ do usually appear from the simplification of included trees in such situations, however, and we are prohibited to discard them.

A further exploitation of the time symmetry is explored in Section 7 where we develop expansions that discard all $\mathcal{O}\left(\varepsilon^{2k\sigma-1}\right)$ terms.

5.1 A 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,

$$\mu_{j,k}(h) := \int_0^h \tilde{B}^k_j(h,\zeta)V(\zeta)\,\mathrm{d}\zeta, \tag{5.8}$$

and integrals over the triangle,

$$\Lambda\left[f\right]_{a,b}(h) := \int_0^h \int_0^\zeta f(h,\zeta,\xi)\left[\partial_x^a V(\zeta)\right]\left[\partial_x^b V(\xi)\right]\,\mathrm{d}\xi\,\mathrm{d}\zeta, \tag{5.9}$$

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

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

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

$$\int_0^h B_j(h,\zeta)\,\mathrm{d}\zeta = 0, \tag{5.10}$$

the term $\mu_{j,1}(h)$ gains an extra power of h and is $\mathcal{O}\left(h^{j+2}\right)$. With this new notation in place, the Magnus expansions $\tilde{\Theta}_3^\varepsilon$ and $\tilde{\Theta}_4^\varepsilon$ can be presented more concisely,

$$\tilde{\Theta}_3^\varepsilon(h) = \overbrace{\mathrm{i}h\varepsilon\partial_x^2 + \mathrm{i}\varepsilon^{-1}\mu_{0,0}(h)}^{\mathcal{O}\left(\varepsilon^{\sigma-1}\right)} + \overbrace{2\left\langle\partial_x\mu_{1,1}(h)\right\rangle_1}^{\mathcal{O}\left(\varepsilon^{3\sigma-1}\right)}, \tag{5.11}$$

$$\tilde{\Theta}_4^\varepsilon(h) = \tilde{\Theta}_3^\varepsilon(h) + \overbrace{\mathrm{i}\varepsilon^{-1}\Lambda\left[\phi\right]_{1,1}(h) - 2\mathrm{i}\varepsilon\left\langle\partial_x^2\mu_{2,1}(h)\right\rangle_2}^{\mathcal{O}\left(\varepsilon^{4\sigma-1}\right)}, \tag{5.12}$$

with

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

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

$$\int_0^h \int_0^\zeta \phi(h,\zeta,\xi)\,\mathrm{d}\xi\,\mathrm{d}\zeta = 0, \tag{5.13}$$

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

6 Symmetric Zassenhaus splittings

Once a suitably truncated Magnus expansion $\tilde{\Theta}_p^\varepsilon(h)$ has been derived, we still need to approximate effectively the exponential $\exp(\tilde{\Theta}_p^\varepsilon(h))u(0)$. This is certainly not easier than the challenge of exponentiating $\mathrm{i}h\varepsilon\partial_x^2 + \mathrm{i}h\varepsilon^{-1}V$ that we face in the case of the time-independent potential since $\tilde{\Theta}_p^\varepsilon(h)$ for $p \geq 2$ always features a term, $\mathrm{i}h\varepsilon\partial_x^2 + \mathrm{i}\varepsilon^{-1}\mu_{0,0}(h)$, with similar structure and size. The matter is complicated further by the inclusion of increasingly complex terms once we consider $p \geq 3$, such as the term $2\left\langle\partial_x\mu_{1,1}(h)\right\rangle_1$ which appears in $\tilde{\Theta}_3^\varepsilon(h)$.

A reasonable way of approximating this exponential is an exponential splitting (Yošida 1990, McLachlan & Quispel 2002, Blanes et al. 2006, Lubich 2008, Jin et al. 2011, Faou 2012). Unlike the Magnus expansion of (Hochbruck & Lubich 2002),

our Magnus expansion is commutator-free and features terms which are smaller in size due to the property of height reduction. However, since we have maintained the expansion in an operatorial form – featuring undiscretised operators – we can resort to a symmetric Zassenhaus splitting for $\exp(\tilde{\Theta}_p^\varepsilon(h))$ along the lines of Bader et al. (2014). This splitting builds on the standard, non-symmetric, Zassenhaus splitting of (Oteo 1991) while correctly accounting for powers of ε and handling undiscretised operators of the form we encounter in the analysis of the Schrödinger equation. This has crucial advantages over conventional splittings (Bader et al. 2014).

The Zassenhaus algorithm provides a neat separation of terms with differing structure and size, each of which is easy to exponentiate separately. It achieves this separation by a recursive application of the symmetric *Baker–Campbell–Hausdorff formula* (usually known in an abbreviated form as the symmetric BCH formula),

$$\mathrm{e}^{\frac{1}{2}X}\mathrm{e}^{Y}\mathrm{e}^{\frac{1}{2}X} = \mathrm{e}^{\mathrm{sBCH}(X,Y)}, \tag{6.1}$$

for X and Y in a Lie algebra $\mathfrak{g}$, where

$$\mathrm{sBCH}(hX,hY) = h(X+Y) - h^3(\tfrac{1}{24}[[Y,X],X] + \tfrac{1}{12}[[Y,X],Y]) + \mathcal{O}\left(h^5\right). \tag{6.2}$$

The expansion (6.2) features terms of the Hall basis (Reutenauer 1993), such as $[[Y,X],X]$ and $[[Y,X],Y]$, and can be computed to an arbitrary power of h using an algorithm from (Casas & Murua 2009). Coefficients and terms of the Hall basis for sufficiently high degree sBCH expansions are also available in a tabular form (Murua 2010). Since (6.1) is palindromic, only odd powers of h feature in the expansion (6.2).

Let us recall the basic principle for the iterative symmetric Zassenhaus splitting (Bader et al. 2014). Our goal is to compute $\mathrm{e}^{\mathcal{W}^{[0]}}$, where $\mathcal{W}^{[0]} = X + Y$. Using the sBCH formula (6.1), we write

$$\mathrm{e}^{\mathcal{W}^{[0]}} = \mathrm{e}^{\frac{1}{2}X}\mathrm{e}^{\mathrm{sBCH}(-X,\mathcal{W}^{[0]})}\mathrm{e}^{\frac{1}{2}X}. \tag{6.3}$$

For $X, Y = \mathcal{O}(h)$, $\mathrm{sBCH}(-X, X+Y) = X + \mathcal{O}\left(h^3\right)$ and thus, we have extracted X from the exponent $\mathcal{W}^{[0]}$ at the cost of correction terms in form of higher-order commutators. However, when accounting in powers of ε, we start with X and Y which might be $\mathcal{O}\left(\varepsilon^0\right)$ or larger, such as the terms $\mathrm{i}h\varepsilon\partial_x^2$ and $\mathrm{i}\varepsilon^{-1}\mu_{0,0}(h)$ under the scaling $0 < \sigma \leq 1$. A similar decrease in the size of correction terms is not observed here till the commutators are simplified in the free Lie algebra where the property of height reduction (Lemma 2) proves crucial.

Assuming that the corrections are decreasing in size, it is then enough to identify the largest terms $W^{[1]}$ in the central exponent $\mathcal{W}^{[1]} = \mathrm{sBCH}(-X, \mathcal{W}^{[0]})$ to continue the iteration until the desired accuracy, say $\mathcal{O}(\varepsilon^r)$, is reached,

$$\mathcal{W}^{[k+1]} = \mathrm{sBCH}(-W^{[k]}, \mathcal{W}^{[k]}) + \mathcal{O}(\varepsilon^r), \quad \mathcal{W}^{[0]} = X + Y, \tag{6.4}$$

where we may discard all $\mathcal{O}(\varepsilon^r)$ or smaller terms at each stage. In this notation, the result after s steps can be written as

$$\exp(X+Y) = \mathrm{e}^{\frac{1}{2}W^{[0]}}\mathrm{e}^{\frac{1}{2}W^{[1]}}\cdots\mathrm{e}^{\frac{1}{2}W^{[s]}}\mathrm{e}^{\mathcal{W}^{[s+1]}}\mathrm{e}^{\frac{1}{2}W^{[s]}}\cdots\mathrm{e}^{\frac{1}{2}W^{[1]}}\mathrm{e}^{\frac{1}{2}W^{[0]}} + \mathcal{O}(\varepsilon^r).$$

We emphasize that, in principle, we are free to choose the elements $W^{[k]}$ that we want to extract. This choice can afford a great deal of flexibility – it could be based on some structural property which allows for trivial exponentiation of $W^{[k]}$ when extracted separately, a small spectral radius which makes the term amenable to effective exponentiation through Krylov subspace methods, a combination of both, or some other criteria. So long as the terms are decreasing in size, the convergence of the procedure is guaranteed. This can lead to many variants of the splitting, some of which could prove to have more favourable properties than others. However, we present the general idea here and do not explore its variants.

All through, we discard terms smaller than our error tolerance – for $\tilde{\Theta}^{\varepsilon}_{2p-1}(h)$ this means discarding terms of size $\mathcal{O}\left(\varepsilon^{(2p+1)\sigma-1}\right)$. For $\tilde{\Theta}^{\varepsilon}_{3}$, where $p=2$, we get a five-stage splitting by starting with $X = \mathrm{i}h\varepsilon\partial_x^2$ and then extracting the largest term at each iteration,

$$\exp\left(\tilde{\Theta}^{\varepsilon}_{3}(h)\right) = \mathrm{e}^{\frac{1}{2}W^{[0]}}\mathrm{e}^{\frac{1}{2}W^{[1]}}\mathrm{e}^{\mathcal{W}^{[2]}}\mathrm{e}^{\frac{1}{2}W^{[1]}}\mathrm{e}^{\frac{1}{2}W^{[0]}} + \mathcal{O}\left(\varepsilon^{5\sigma-1}\right), \tag{6.5}$$

where

$$\begin{aligned}
W^{[0]} &= \mathrm{i}h\varepsilon\partial_x^2 \quad &&= \mathcal{O}\left(\varepsilon^{\sigma-1}\right),\\
W^{[1]} &= \mathrm{i}\varepsilon^{-1}\mu_{0,0}(h) \quad &&= \mathcal{O}\left(\varepsilon^{\sigma-1}\right),\\
\mathcal{W}^{[2]} &= \tfrac{1}{6}\mathrm{i}h\varepsilon^{-1}\left(\partial_x\mu_{0,0}(h)\right)^2 + 2\left\langle\partial_x\mu_{1,1}(h)\right\rangle_1 - \tfrac{1}{6}\mathrm{i}h^2\varepsilon\left\langle\partial_x^2\mu_{0,0}(h)\right\rangle_2 = \mathcal{O}\left(\varepsilon^{3\sigma-1}\right).
\end{aligned}$$

As a quick sanity check, we do arrive at the standard symmetric Zassenhaus splitting (Bader et al. 2014) for time-independent potentials, $V(x,t) := V(x)$.

It is only by this stage – having arrived at an asymptotic splitting expressed in operatorial terms – that we start considering discretisation issues. Considerations of spatial and temporal discretisation (in the form of approximation through quadrature) are entirely independent here and one may proceed to address them in any order.

6.1 Evaluation of integrals

At first, we consider the problem of evaluating the integrals appearing in our splitting: $\mu_{0,0}(h)$ and $\mu_{1,1}(h)$. The integral

$$\mu_{0,0}(h) = \int_0^h V(\zeta)\,\mathrm{d}\zeta$$

appears once in $W^{[1]}$ and twice in $\mathcal{W}^{[2]}$. Since we have already committed an $\mathcal{O}\left(\varepsilon^{5\sigma-1}\right)$ error in the splitting, we need not approximate the integrals to a greater degree of accuracy. In the term $\frac{1}{6}\mathrm{i}h^2\varepsilon\left\langle\partial_x^2\mu_{0,0}(h)\right\rangle_2$, we need an $\mathcal{O}\left(\varepsilon^{3\sigma}\right)$ or $\mathcal{O}\left(h^3\right)$ approximation of $\mu_{0,0}(h)$ due to the presence of the $h^2\varepsilon$ scalar factor and since $\langle f\rangle_2$ is $\mathcal{O}\left(\varepsilon^{-2}\right)$ for $f = \mathcal{O}\left(\varepsilon^0\right)$. However, $\mathcal{O}\left(h^4\right)$ and $\mathcal{O}\left(h^5\right)$ approximations are needed for its other occurrences in $\mathcal{W}^{[2]}$ and $W^{[1]}$, respectively. The only occurrence of $\mu_{1,1}(h)$,

$$\mu_{1,1}(h) = \int_0^h \left(\zeta - \tfrac{h}{2}\right) V(\zeta)\,\mathrm{d}\zeta,$$

demands an $\mathcal{O}\left(h^5\right)$ approximation. Thus, it suffices to evaluate $\mathcal{O}\left(h^5\right)$ accuracy approximations for the integrals $\mu_{0,0}(h)$ and $\mu_{1,1}(h)$ once per time step.

In the standard setting, where $V(h)$ is $\mathcal{O}\left(h^0\right)$ and is sufficiently smooth in the temporal domain, we may effectively approximate the integrals using two Gauss–Legendre knots $t_k := \frac{h}{2}(1 + k/\sqrt{3})$, $k = -1, 1$, and weights $w_k = \frac{h}{2}$ (Davis & Rabinowitz 1984),

$$\mu_{0,0}(h) = \left(\frac{V(t_1) + V(t_{-1})}{2}\right) h + \mathcal{O}\left(h^5\right),$$

and

$$\mu_{1,1}(h) = \left(\frac{V(t_1) - V(t_{-1})}{4\sqrt{3}}\right) h^2 + \mathcal{O}\left(h^5\right).$$

These turn out to be centred finite difference approximations of $\partial_t^j V(t)$ at $t = \frac{h}{2}$ for $j = 0, 1$ on the grid $\frac{h}{2}(1 + k/\sqrt{3}), k = -1, 0, 1$. This is not altogether surprising.

Instead of preserving the integrals till the very end, as we have chosen to do here, it is possible to start directly from the results of (Munthe–Kaas & Owren 1999) where the Magnus expansion is developed with integrals replaced by quadrature at the outset. This approach is equivalent to the Taylor series development of the integrand by Blanes, Casas, Oteo & Ros (2009). When discretising with Gauss–Legendre knots in either case, we arrive at Magnus expansions and Zassenhaus splittings featuring the above approximations once more. This alternative approach, where we commence our analysis from an integral-free approximation of the Magnus expansion, is under development in parallel and will feature in another publication.

While the approach presented here requires us to carry all integrals along in an undiscretised form, and is thus considerably more involved, it is also more flexible. Little has been assumed about the regularity of V in the temporal domain so far. It is generally accepted that integrals are to be preferred over derivatives – they are particularly helpful in the case of functions with low regularity and in the case of highly oscillatory functions. Here our decision of not replacing the integrals with Taylor expansion at the outset affords us the ability to effectively handle such cases.

It might be possible to evaluate the exact integral for $\mu_{0,0}(h)$ and $\mu_{1,1}(h)$, for instance, if an analytic expression for V is available. This is also true for potentials of the form $V(x,t) = f(t)V(x)$ where an analytic expression for $f(t)$ is available. If $f(t)$ is a highly oscillatory envelope, oscillating around the origin, there can be a further decrease in size of integrals involved which can benefit us greatly.

Where an exact integral is not available, integrals featuring highly oscillatory integrands of the form

$$\int_0^1 f(t)\, \mathrm{e}^{\mathrm{i}\omega g(t)}\, \mathrm{d}t$$

can be approximated effectively using the Filon method (Iserles & Nørsett 2005). The time-varying potential L_ω introduced earlier,

$$L_\omega(x,t) = 2\rho(t)\sin((x - 5t)\pi\omega), \tag{1.4}$$

for instance, can be integrated in this manner. The significance of such quadratures increases when we start considering very highly oscillatory potentials with oscillations of the order of ε^{-1} or larger, as might be expected in some applications of quantum control.

6.2 Spatial discretisation

In the numerical solution of the Schrödinger equation it is common to resort to *spectral* methods for spatial discretisation (Jin et al. 2011, Faou 2012). We follow the spectral collocation or pseudo-spectral approach (Fornberg 1998, Hesthaven, Gottlieb & Gottlieb 2007), where the idea is to interpolate the solution at nodal values using a trigonometric polynomial. Since it is compelling in the presence of periodic boundary conditions to use equispaced points, the unknowns are $u_n \approx u(n/(N+\frac{1}{2}))$, $|n| \leq N$, where $M = 2N+1$. The main appeal of spectral methods is that they exhibit *spectral convergence:* for sufficiently large M the error decays faster than $\mathcal{O}(M^{-\alpha}) = \mathcal{O}(\varepsilon^{\alpha})$ for any $\alpha > 0$. In classical terms, the method is of an infinite order.

The wavefunction $u \in C_p^{\infty}([-1,1];\mathbb{C})$ is discretised as a vector $\boldsymbol{u} \in \mathbb{C}^M$. We write $\mathcal{D}_{a_n}$ to denote the diagonal $M \times M$ matrix whose diagonal entries form the sequence $\{a_n\}_{n=-N}^{N}$. We write $\mathcal{D}_f$ to denote the diagonal matrix corresponding to pointwise multiplication by f on the grid $\{x_n\}_{n=-N}^{N}$, $x_n = n/(N+\frac{1}{2})$, where the underlying sequence is $\{f(x_n)\}_{n=-N}^{N}$. The differential operator ∂_x is discretised as a circulant matrix $\mathcal{K}$, which is diagonalisable using the Fast Fourier Transform matrix $\mathcal{F}$,

$$\partial_x \rightsquigarrow \mathcal{K} = \mathcal{F}^{-1}\mathcal{D}_{\mathrm{i}n\pi}\mathcal{F}.$$

The computation of $\mathcal{K}\boldsymbol{u}$, which involves two FFTs, costs $\mathcal{O}(M \log M)$ operations, while $D_f\boldsymbol{u}$ costs $\mathcal{O}(M)$ operations as it can be evaluated in a pointwise fashion.

6.3 Approximation of exponentials

In the time-stepping procedure (2.3), we are interested in approximating $\exp(\Theta(h))u(0)$. For the splitting (6.5), this breaks down into a sequence of matrix-vector products of the form $\exp(\boldsymbol{W})\boldsymbol{u}$ which need be approximated.

We note that $G(\mathcal{D}_f) = \mathcal{D}_{G(f)}$ for any analytic operator G, while the unitarity of the Fourier transform implies that $G(\mathcal{K}) = \mathcal{F}^{-1}\mathcal{D}_{G(\mathrm{i}m\pi)}\mathcal{F}$. Consequently, the exponential of $W^{[0]} = \mathrm{i}h\varepsilon\partial_x^2$ is evaluated using two FFTs,

$$\mathrm{e}^{\frac{1}{2}W^{[0]}}u \rightsquigarrow \mathcal{F}^{-1}\mathcal{D}_{\exp(-\mathrm{i}h\varepsilon n^2\pi^2/2)}\mathcal{F}\boldsymbol{u},$$

in $\mathcal{O}(M \log M)$ operations, and $W^{[1]}$ is exponentiated trivially in $\mathcal{O}(M)$ operations,

$$\mathrm{e}^{\frac{1}{2}W^{[1]}}u = \exp\left(\tfrac{1}{2}\mathrm{i}\varepsilon^{-1}\mu_{0,0}(h)\right)u \rightsquigarrow \mathcal{D}_{\exp(\frac{1}{2}\mathrm{i}\varepsilon^{-1}\mu_{0,0}(h))}\boldsymbol{u}.$$

$W^{[0]}$ and $W^{[1]}$, which happen to be the largest of the exponents, have been exponentiated directly. Since both calculations are exact (up to machine accuracy), unitarity is maintained. No direct way of exponentiating the remaining exponent, $\mathcal{W}^{[2]}$, presents itself in general. To deal with this we use Lanczos iterations, which is a *Krylov subspace method.* Such methods have undergone many enhancements since the pioneering work of Tal Ezer & Kosloff (1984): in the current paper we adopt the approach in (Gallopoulos & Saad 1992).

Given an $M \times M$ matrix $\mathcal{A}$ and $\boldsymbol{v} \in \mathbb{C}^M$, the mth Krylov subspace is

$$\boldsymbol{K}_m(\mathcal{A},\boldsymbol{v}) = \mathrm{span}\,\{\boldsymbol{v}, \mathcal{A}\boldsymbol{v}, \mathcal{A}^2\boldsymbol{v}, \ldots, \mathcal{A}^{m-1}\boldsymbol{v}\}, \qquad m \in \mathbb{N}.$$

It is well known that $\dim \boldsymbol{K}_{m-1}(\mathcal{A},\boldsymbol{v}) \le \dim \boldsymbol{K}_m(\mathcal{A},\boldsymbol{v}) \le \min\{m,M\}$ and we refer to (Golub & Van Loan 1996) for other properties of Krylov subspaces. The main idea is to approximate

$$\mathrm{e}^{\mathcal{A}}\boldsymbol{v} \approx \mathcal{V}_m \mathrm{e}^{\mathcal{H}_m}\mathcal{V}_m^*\boldsymbol{v}, \tag{6.6}$$

where $\mathcal{V}_m$ and $\mathcal{H}_m$ are $M\times m$ and $m\times m$ respectively and $m \ll M$. In addition, the columns of $\mathcal{V}_m$ are orthonormal vectors which form a basis of $\boldsymbol{K}_m(\mathcal{A},\boldsymbol{v})$, while $\mathcal{H}_m$ is upper Hessenberg.

The matrices $\mathcal{V}_m$ and $\mathcal{H}_m$ are generated by *Lanczos iterations*

Lanczos iterations

$h_{0,1} = 0$
$\boldsymbol{v}_0 = \boldsymbol{0}$
$\boldsymbol{v}_1 = \boldsymbol{v}/\|\boldsymbol{v}\|_2$
for $j = 1,\dots,m$ **do**
  $\boldsymbol{t} = \mathcal{A}\boldsymbol{v}_j$
  $h_{j,j} = \boldsymbol{v}_j^*\boldsymbol{t}$
  $\boldsymbol{t} = \boldsymbol{t} - h_{j,j}\boldsymbol{v}_j - h_{j-1,j}\boldsymbol{v}_{j-1}$
  $h_{j+1,j} = \|\boldsymbol{t}\|_2, \qquad h_{j,j+1} = -h_{j+1,j}$
  $\boldsymbol{v}_{j+1} = \boldsymbol{t}/h_{j+1,j}$
end for

(Golub & Van Loan 1996, Gallopoulos & Saad 1992). Note that, once $\mathcal{A}\in\mathfrak{u}_M(\mathbb{C})$, where $\mathfrak{u}_M(\mathbb{C})$ is the Lie algebra of $M\times M$ skew-Hermitian matrices, it follows that $\mathcal{H}_m \in \mathfrak{u}_m(\mathbb{C})$. Therefore, the columns of $\mathcal{V}_m$ being orthonormal, the ℓ_2 norm is conserved. Moreover, since $\mathcal{V}_m^*\boldsymbol{v} = \|\boldsymbol{v}\|_2\boldsymbol{e}_1$, where $\boldsymbol{e}_1\in\mathbb{C}^m$ is the first unit vector, it follows that $\mathrm{e}^{\mathcal{H}_m}\mathcal{V}_m^*\boldsymbol{v}$ is merely the first column of $\mathrm{e}^{\mathcal{H}_m}$, scaled by $\|\boldsymbol{v}\|_2$. To compute the approximation (6.6) we thus need to evaluate a *small* exponential and calculate a single matrix-vector product.

For small values of m, once $\mathcal{H}_m$ and $\mathcal{V}_m$ are available, approximating the exponential is very inexpensive. The computational cost is, therefore, largely governed by the cost of the m iterations, each involving a matrix-vector product of the form $\mathcal{A}\boldsymbol{v}_j$.

The only exponent, $\mathcal{W}^{[2]}$, for which we resort to Lanczos iterations in the splitting (6.5) features Jordan terms of the form $\langle f\rangle_k = (f\circ\partial_x^k + \partial_x^k\circ f)/2$. These are discretised in a straightforward way,

$$\langle f\rangle_k \rightsquigarrow \frac{1}{2}(\mathcal{D}_f\mathcal{K}^k + \mathcal{K}^k\mathcal{D}_f) = \frac{1}{2}\left(\mathcal{D}_f\mathcal{F}^{-1}\mathcal{D}_{(\mathrm{i}n\pi)^k}\mathcal{F} + \mathcal{F}^{-1}\mathcal{D}_{(\mathrm{i}n\pi)^k}\mathcal{F}\mathcal{D}_f\right),$$

and the cost of approximating $\langle f\rangle_k u$ by $\frac{1}{2}\left(\mathcal{D}_f\mathcal{F}^{-1}\mathcal{D}_{(\mathrm{i}n\pi)^k}\mathcal{F}\boldsymbol{u} + \mathcal{F}^{-1}\mathcal{D}_{(\mathrm{i}n\pi)^k}\mathcal{F}\mathcal{D}_f\boldsymbol{u}\right)$ is dominated by the $\mathcal{O}(M\log M)$ cost of the four FFTs. Approximating $\mathcal{W}^{[2]}\boldsymbol{v}_j$ in every Lanczos iteration is an $\mathcal{O}(M\log M)$ affair, therefore, bringing the overall cost to $\mathcal{O}(mM\log M)$ operations.

The terms in $\mathcal{W}^{[2]}$ feature expressions of the form $\partial_x\mu_{0,0}(h)$, $\partial_x\mu_{1,1}(h)$ and $\partial_x^2\mu_{0,0}(h)$ which might be available in closed form. However, in general these can be evaluated through numerical differentiation if $\mu_{0,0}(h)$ and $\mu_{1,1}(h)$, or their suitable approximations, are available. $\partial_x^2\mu_{0,0}(h)$, for instance, can be approximated by the vector $\mathcal{K}^2\boldsymbol{\mu}_{0,0}(h)$. We note that the degree of accuracy required in the differentiation here

is considerably smaller than the spectral accuracy of spectral-collocation differentiation matrices. We may, instead, use centred finite-difference differentiation matrices of varying accuracy (depending on the accuracy requirement per term) to achieve the same feat at an $\mathcal{O}(M)$ cost.

The question of an appropriate value of m is answered by the inequality

$$\|\mathrm{e}^{\mathcal{A}}\boldsymbol{v} - \mathcal{V}_m \mathrm{e}^{\mathcal{H}_m}\mathcal{V}_m^*\boldsymbol{v}\|_2 \leq 12\mathrm{e}^{-\rho^2/(4m)}\left(\frac{\mathrm{e}\rho}{2m}\right)^m, \qquad m \geq \rho, \tag{6.7}$$

where $\rho = \rho(\mathcal{A})$ is the spectral radius of $\mathcal{A}$ (Hochbruck & Lubich 1997). Under the scaling choice $\sigma = 1$, we know that $\mathcal{W}^{[2]} = \mathcal{O}(\varepsilon^2)$ and assume, with very minor loss of generality, that $\rho(\mathcal{W}^{[2]}) \leq c\varepsilon^2$ for some $c > 0$. We thus deduce from (6.7) that

$$\|\mathrm{e}^{\mathcal{W}^{[2]}}\boldsymbol{v} - \mathcal{V}_m \mathrm{e}^{\mathcal{H}_m}\mathcal{V}_m^*\boldsymbol{v}\|_2 \leq 12\left(\frac{\mathrm{e}c}{2m}\right)^m \varepsilon^{2m}, \qquad m \geq \rho,$$

and $m = 2$ is sufficient to reduce the error to $\mathcal{O}(\varepsilon^4)$, in line with the error of our symmetric Zassenhaus algorithm. The number of iterations required do change with the choice of σ. For $\sigma = 1/2$, for instance, under which $\mathcal{W}^{[2]}$ is $\mathcal{O}(\varepsilon^{1/2})$, we require $m = 3$ for achieving the $\mathcal{O}(\varepsilon^{3/2})$ accuracy of our splitting.

A critical stage is reached at $\sigma = 1/3$, where the term $\mathcal{W}^{[2]}$ becomes $\mathcal{O}(\varepsilon^0)$ and no longer decreases in size with ε. Beyond this, the exponent grows in size with decreasing ε. Thus we are forced to place a limit on the time-step scaling and restrict ourselves to $1/3 < \sigma \leq 1$. This restriction, nevertheless, is more generous than the $\sigma \geq 1$ restriction of (Hochbruck & Lubich 2002).

6.4 Numerical experiments

For numerical experiments we recall the setting discussed in the introduction: we have a wave-packet (1.2) with $x_0 = -0.3$ and $\delta = 1.22 \times 10^{-4}$, as before, sitting in the double-well potential (1.3) (Figure 1.1) and excited by the time-varying field (1.4),

$$\begin{aligned} u(0) &= u_0, \\ V_{100}(x,t) &= V_0 + L_{100}(x,t), \end{aligned}$$

with periodic boundaries imposed at $x = \pm 1$. The errors presented are the global errors in $u(x,T)$ where $T = 0.1$ is the final time.

Under the scaling $\sigma = 1$, we commit a local error of $\mathcal{O}(\varepsilon^4)$ per time step in the splitting scheme (6.5). The precise scaling used in our numerical experiments is

$$M \sim 5\varepsilon^{-1}, \quad h \sim \varepsilon.$$

Since the number of time steps is $\mathcal{O}(\varepsilon^{-\sigma})$, the global error is $\mathcal{O}(\varepsilon^3)$.

Our analysis has been in the context of the L_2 inner product and the corresponding norm which, upon discretisation, translates to an ℓ_2 norm (scaled by a factor of $\sqrt{2/M}$). Where L_∞ error is of greater interest, it should be noted that $\|\boldsymbol{v}\|_{\ell_\infty} \leq \sqrt{M/2}\|\boldsymbol{v}\|_{\ell_2}$ and consequently we may expect the global L_∞ error to be $\mathcal{O}(\varepsilon^{5/2})$. This is indeed seen to be the case through numerical experiments in Figure 6.3.

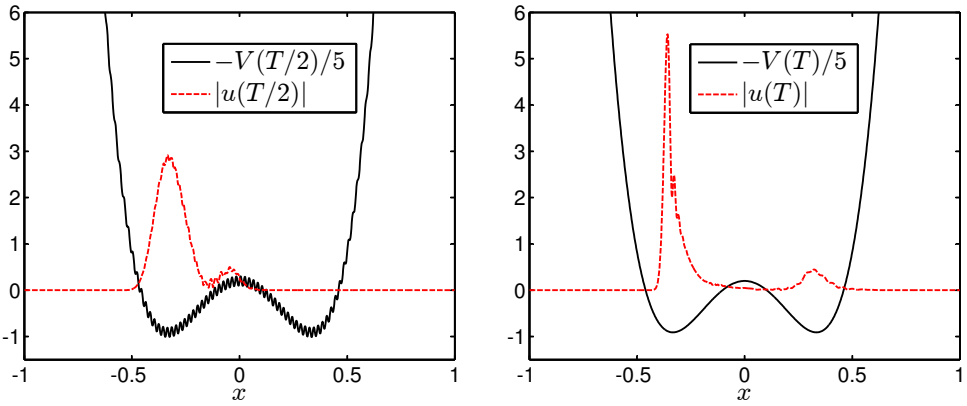


Figure 6.2: Wave packet $u(x,t)$ under the influence of $V_{100}(x,t) = V_0 + L_{100}(x,t)$ at $t = T/2$ (left) and at $t = T$ (right). For consistency with physical interpretations we depict the negative potential, scaling it down by a factor of five for ease of illustration.

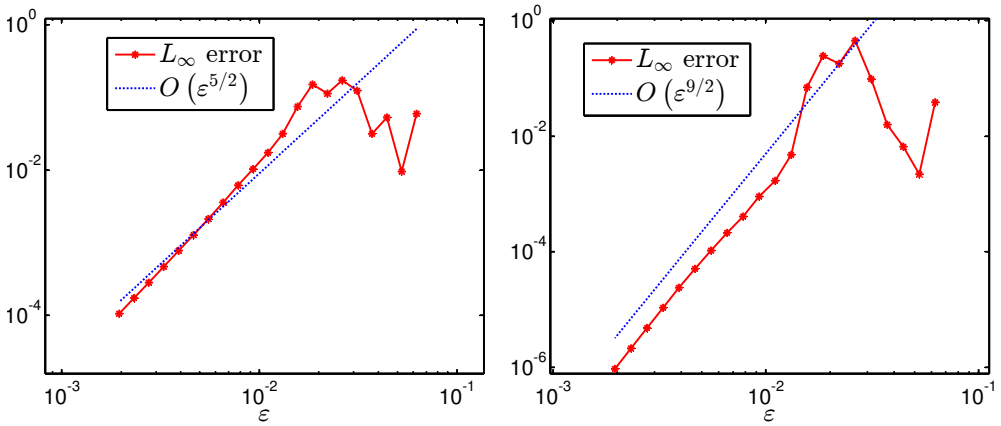


Figure 6.3: Global L_∞ errors of the Zassenhaus splittings at $T = 0.1$ under the scaling $\sigma = 1$: (left) error for the splitting (6.5) is $\mathcal{O}\left(\varepsilon^{5/2}\right)$, (right) error for the higher order splitting (7.12) is $\mathcal{O}\left(\varepsilon^{9/2}\right)$.

7 Towards higher order splittings

Arbitrarily high order Zassenhaus splittings for the Schrödinger equation (1.1) can be obtained by starting from a high order Magnus expansion. For a Zassenhaus splitting featuring an error of $\mathcal{O}\left(\varepsilon^{7\sigma-1}\right)$, for instance, we need to consider the Magnus expansion

$\tilde{\Theta}_5^\varepsilon$ which is derived by simplifying trees in Θ_5 and discarding all $\mathcal{O}\left(\varepsilon^{7\sigma-1}\right)$ terms,

$$\begin{aligned}\tilde{\Theta}_5^\varepsilon(h) = &\overbrace{\mathrm{i}h\varepsilon\partial_x^2 + \mathrm{i}\varepsilon^{-1}\mu_{0,0}(h)}^{\mathcal{O}\left(\varepsilon^{\sigma-1}\right)} + \overbrace{2\left\langle\partial_x\mu_{1,1}(h)\right\rangle_1}^{\mathcal{O}\left(\varepsilon^{3\sigma-1}\right)} + \overbrace{\mathrm{i}\varepsilon^{-1}\Lambda\left[\phi\right]_{1,1}(h) - 2\mathrm{i}\varepsilon\left\langle\partial_x^2\mu_{2,1}(h)\right\rangle_2}^{\mathcal{O}\left(\varepsilon^{4\sigma-1}\right)}\\ &+\overbrace{\tfrac{1}{6}\left\langle\Lambda\left[\psi_1\right]_{1,2}(h) + \Lambda\left[\psi_2\right]_{2,1}(h)\right\rangle_1}^{\mathcal{O}\left(\varepsilon^{4\sigma-1}\right)} + \overbrace{\tfrac{1}{6}\left\langle\Lambda\left[\theta_1\right]_{1,2}(h) + \Lambda\left[\theta_2\right]_{2,1}(h)\right\rangle_1}^{\mathcal{O}\left(\varepsilon^{5\sigma-1}\right)}\\ &-\overbrace{\tfrac{4}{3}\varepsilon^2\left\langle\partial_x^3\mu_{1,3}(h)\right\rangle_3}^{\mathcal{O}\left(\varepsilon^{5\sigma-1}\right)} - \overbrace{\tfrac{1}{4}\mathrm{i}\varepsilon\partial_x^4\mu_{2,1}(h)}^{\mathcal{O}\left(\varepsilon^{6\sigma-1}\right)} = \Theta(h) + \mathcal{O}\left(\varepsilon^{7\sigma-1}\right),\end{aligned} \tag{7.1}$$

where

$$\begin{aligned}\psi_1(h,\zeta,\xi) &:= h^2 - 4h\xi + 2\zeta\xi,\\ \psi_2(h,\zeta,\xi) &:= (h-2\zeta)^2 - 2\zeta\xi,\\ \theta_1(h,\zeta,\xi) &:= h^2 - 6h\zeta + 6h\xi + 6\zeta\xi + 3\zeta^2 - 12\xi^2,\\ \theta_2(h,\zeta,\xi) &:= h^2 - 6h\zeta + 6h\xi - 6\zeta\xi + 5\zeta^2.\end{aligned} \tag{7.2}$$

Integrals of θ_j vanish over the triangle,

$$\int_0^h\int_0^\zeta \theta_j(h,\zeta,\xi)\,\mathrm{d}\xi\,\mathrm{d}\zeta = 0, \quad j=1,2, \tag{7.3}$$

lending an extra power of h to the functionals where θ_js appear. No similar observation about ψ_js can be made and, although similar in many regards, functionals featuring them ought not be combined with the corresponding ones featuring θ_js by this stage.

When we commence the Zassenhaus splitting procedure with the truncated Magnus expansion $\tilde{\Theta}_5^\varepsilon$ featuring terms of sizes $\mathcal{O}\left(\varepsilon^{k\sigma-1}\right)$, $k=1,3,4,5,6$, the resulting exponential splitting will continue to feature terms of all these sizes. The expansion $\tilde{\Theta}_7^\varepsilon$ features $\mathcal{O}\left(\varepsilon^{k\sigma-1}\right)$ terms with $k=1,3,4,5,6,7,8$, and its Zassenhaus expansion, once again, retains all such terms. This is suboptimal – the time-symmetric nature of the power-truncated Magnus expansion Θ_p implies that it should be possible to expand $\Theta_p^\varepsilon(h)$ and, therefore, $\tilde{\Theta}_p^\varepsilon(h)$ solely in odd powers of h for any choice of the potential, V. A Zassenhaus splitting commencing from such an odd-powered expansion will never introduce even powers of h since underlying the procedure is a recursive application of the symmetric BCH which features only odd-grade commutators.

To examine the time symmetry of the Magnus expansion, we revisit (2.2),

$$u(t+h) = \mathrm{e}^{\Theta(t+h,t)}u(t), \tag{2.2}$$

where the exponential, $\exp(\Theta(t+h,t))$, is the evolution operator from t to $t+h$. As we had remarked earlier, $\Theta(t+h,t)$ can be easily recovered from $\Theta(h,0)$ (shortened to $\Theta(h)$) by substituting all occurrences of $A(\zeta)$ with $A(t+\zeta)$.

We define $t_{1/2} = t + h/2$ as the midpoint of the interval $[t, t+h]$ and rewrite (2.2) as

$$u(t_{1/2}+\tfrac{h}{2}) = \mathrm{e}^{\Theta(t_{1/2}+\frac{h}{2},\,t_{1/2}-\frac{h}{2})}u(t_{1/2}-\tfrac{h}{2}). \tag{7.4}$$

Since $\Theta(t_{1/2}-\frac{h}{2},t_{1/2}+\frac{h}{2})$ is the evolution operator from $t_{1/2}+\frac{h}{2}$ to $t_{1/2}-\frac{h}{2}$ (going backward in time by length h), we also have

$$u(t_{1/2}-\tfrac{h}{2}) = \mathrm{e}^{\Theta(t_{1/2}-\frac{h}{2},t_{1/2}+\frac{h}{2})}u(t_{1/2}+\tfrac{h}{2}). \tag{7.5}$$

Combining the two, we find that $\Theta(t_{1/2}-\frac{h}{2},t_{1/2}+\frac{h}{2}) = -\Theta(t_{1/2}+\frac{h}{2},t_{1/2}-\frac{h}{2})$, so that Θ is odd in h around the midpoint $t_{1/2}$. Similarly, the power-truncated Magnus expansions $\Theta_p^\varepsilon(t_{1/2}+\frac{h}{2},t_{1/2}-\frac{h}{2})$ are also odd in h around $t_{1/2}$. If we expand Θ_p^ε in powers of h around $t_{1/2}$, therefore, we should only get odd powers of h.

The expansion $\Theta_p^\varepsilon(t_{1/2}+\frac{h}{2},t_{1/2}-\frac{h}{2}) = \Theta(t+h,t)$ can be obtained from $\Theta_p^\varepsilon(h) = \Theta_p^\varepsilon(h,0)$ by substituting all occurrences of $V(\zeta)$ by $V(t_{1/2}-\frac{h}{2}+\zeta)$. Keeping the odd nature of the expansion about $t_{1/2}$ in mind, we shift the origin to $t_{1/2}$ by defining

$$W(\zeta) := V(t_{1/2}+\zeta),$$

whereby we need to substitute $V(\zeta)$ with $W(\zeta-\frac{h}{2})$. To arrive at the desired expansion of $\Theta_p^\varepsilon(t+h,t)$, therefore, we only need to substitute occurrences of $\mu_{j,k}(h)$ and $\Lambda[f]_{a,b}(h)$ with the new definitions,

$$\mu_{j,k}(h) := \int_0^h \tilde{B}_j^k(h,\zeta)\, W\left(\zeta-\frac{h}{2}\right)\mathrm{d}\zeta, \tag{7.6}$$

$$\Lambda[f]_{a,b}(h) := \int_0^h\int_0^\zeta f(h,\zeta,\xi)\left[\partial_x^a W\left(\zeta-\frac{h}{2}\right)\ \partial_x^b W\left(\xi-\frac{h}{2}\right)\right]\mathrm{d}\xi\,\mathrm{d}\zeta. \tag{7.7}$$

Since we have shifted the origin to $t_{1/2}$, all odd and even components are to be understood with respect to 0 from this point onwards. This makes identification of the odd components of the Magnus expansion simpler, assuming that the odd and even components of W can be found.

A multivariate function F is said to be odd if $F(-\boldsymbol{\zeta}) = -F(\boldsymbol{\zeta})$ and even if $F(-\boldsymbol{\zeta}) = F(\boldsymbol{\zeta})$. The odd and even components, F^o and F^e, of a multivariate function F are defined as

$$F^o(\boldsymbol{\zeta}) := \tfrac{1}{2}\left[F(\boldsymbol{\zeta})-F(-\boldsymbol{\zeta})\right]$$

and

$$F^e(\boldsymbol{\zeta}) := \tfrac{1}{2}\left[F(\boldsymbol{\zeta})+F(-\boldsymbol{\zeta})\right],$$

respectively. It follows that the odd and even components of a product of two multivariate functions are

$$(F(\boldsymbol{\zeta})G(\boldsymbol{\zeta}))^o = F^e(\boldsymbol{\zeta})G^o(\boldsymbol{\zeta}) + F^o(\boldsymbol{\zeta})G^e(\boldsymbol{\zeta})$$

and

$$(F(\boldsymbol{\zeta})G(\boldsymbol{\zeta}))^e = F^e(\boldsymbol{\zeta})G^e(\boldsymbol{\zeta}) + F^o(\boldsymbol{\zeta})G^o(\boldsymbol{\zeta}),$$

respectively. We stress that the odd-ness or even-ness of a multivariate function is considered with respect to all its parameters and therefore changes with context. In the integration identities,

$$\left(\int_0^s F(r,\boldsymbol{\zeta})\,\mathrm{d}r\right)^o = \int_0^s F^e(r,\boldsymbol{\zeta})\,\mathrm{d}r, \tag{7.8}$$

and

$$\left(\int_0^s F(r,\boldsymbol{\zeta})\,\mathrm{d}r\right)^e = \int_0^s F^o(r,\boldsymbol{\zeta})\,\mathrm{d}r, \tag{7.9}$$

for instance, these are with respect to $(s,\boldsymbol{\zeta})$ on the left hand side but with respect to $(r,\boldsymbol{\zeta})$ on the right hand side.

Extending the new definitions of μ and Λ (7.6, 7.7), we define

$$\mu_{\star,j,k}(h) := \int_0^h \tilde{B}_j^k(h,\zeta)\,W^\star\left(\zeta-\frac{h}{2}\right)\,\mathrm{d}\zeta, \quad \star\in\{e,o\}, \tag{7.10}$$

and

$$\Lambda\left[f\right]_{\star,a,b}(h) := \int_0^h\int_0^\zeta f(h,\zeta,\xi)\left[\partial_x^a W\left(\zeta-\frac{h}{2}\right)\;\partial_x^b W\left(\xi-\frac{h}{2}\right)\right]^\star \mathrm{d}\xi\,\mathrm{d}\zeta, \quad \star\in\{e,o\}. \tag{7.11}$$

The even and odd components of previously defined terms $\mu_{j,k}(h)$ and $\Lambda\left[f\right]_{a,b}(h)$ can be expressed in terms of these new definitions. It should be obvious that $\tilde{B}_j(h,\zeta)$ is odd in (h,ζ) for odd values of j and even for even values. Consequently,

$$\begin{aligned}
(\mu_{j,k}(h))^o &= \int_0^h \left(\tilde{B}_j^k(h,\zeta)\,W\left(\zeta-\frac{h}{2}\right)\right)^e \mathrm{d}\zeta \\
&= \int_0^h \left(\tilde{B}_j^k(h,\zeta)\right)^e W^e\left(\zeta-\frac{h}{2}\right) + \left(\tilde{B}_j^k(h,\zeta)\right)^o W^o\left(\zeta-\frac{h}{2}\right)\,\mathrm{d}\zeta \\
&= \begin{cases} \mu_{e,j,k}(h) & \text{if } jk \text{ is even,} \\ \\ \mu_{o,j,k}(h) & \text{if } jk \text{ is odd.} \end{cases}
\end{aligned}$$

In particular, $\mu_{0,0}(h)^o = \mu_{e,0,0}(h) = \int_0^h W^e\left(\zeta-\frac{h}{2}\right)\,\mathrm{d}\zeta$. Note carefully the difference of usage in subscript and superscript. The odd part of $\Lambda\left[f\right]_{a,b}(h)$ is

$$\begin{aligned}
\left(\Lambda\left[f\right]_{a,b}(h)\right)^o &= \int_0^h\int_0^\zeta \left\{f(h,\zeta,\xi)\left[\partial_x^a W\left(\zeta-\frac{h}{2}\right)\;\partial_x^b W\left(\xi-\frac{h}{2}\right)\right]\right\}^o \mathrm{d}\xi\,\mathrm{d}\zeta \\
&= \int_0^h\int_0^\zeta f^o(h,\zeta,\xi)\left[\partial_x^a W\left(\zeta-\frac{h}{2}\right)\;\partial_x^b W\left(\xi-\frac{h}{2}\right)\right]^e \mathrm{d}\xi\,\mathrm{d}\zeta \\
&\quad + \int_0^h\int_0^\zeta f^e(h,\zeta,\xi)\left[\partial_x^a W\left(\zeta-\frac{h}{2}\right)\;\partial_x^b W\left(\xi-\frac{h}{2}\right)\right]^o \mathrm{d}\xi\,\mathrm{d}\zeta \\
&= \Lambda\left[f^o\right]_{e,a,b}(h) + \Lambda\left[f^e\right]_{o,a,b}(h).
\end{aligned}$$

For an odd function such as $\phi(h,\zeta,\xi) := \zeta-\xi-\frac{h}{3}$, this reduces to

$$\left(\Lambda\left[\phi\right]_{a,b}(h)\right)^o = \Lambda\left[\phi\right]_{e,a,b}(h).$$

The even parts $\mu_{j,k}(h)^e$ and $\Lambda\left[f\right]_{a,b}(h)^e$ can be deduced analogously.

The time symmetry of power truncated Magnus expansions ensures that $\Theta_p^\varepsilon(h)$ and $\tilde\Theta_p^\varepsilon(h)$ are odd around the origin so that the even part $\tilde\Theta_p^\varepsilon(h)^e$ vanishes, leaving $\tilde\Theta_p^\varepsilon(h)=\tilde\Theta_p^\varepsilon(h)^o$. Even terms in the Magnus expansion may, therefore, be discarded without further analysis. For instance, we conclude that

$$\begin{aligned}\tilde\Theta_4^\varepsilon(h) &= \mathrm{i}h\varepsilon\partial_x^2+\mathrm{i}\varepsilon^{-1}\left(\mu_{0,0}(h)\right)^o+2\left\langle\partial_x\left(\mu_{1,1}(h)\right)^o\right\rangle_1\\ &\quad+\mathrm{i}\varepsilon^{-1}\Big(\Lambda\left[\phi\right]_{1,1}(h)\Big)^o-2\mathrm{i}\varepsilon\left\langle\partial_x^2\left(\mu_{2,1}(h)\right)^o\right\rangle_2\\ &= \mathrm{i}h\varepsilon\partial_x^2+\mathrm{i}\varepsilon^{-1}\mu_{e,0,0}(h)+2\left\langle\partial_x\mu_{o,1,1}(h)\right\rangle_1\\ &\quad+\mathrm{i}\varepsilon^{-1}\Lambda\left[\phi\right]_{e,1,1}(h)-2\mathrm{i}\varepsilon\left\langle\partial_x^2\mu_{e,2,1}(h)\right\rangle_2.\end{aligned}$$

For an analytic potential,

$$W\left(\zeta-\frac{h}{2}\right)=\sum_{k=0}^\infty\frac{W^{(k)}(0)}{k!}\left(\zeta-\frac{h}{2}\right)^k,$$

we expect a gain of a single power of h in $\mu_{j,1}(h)$ which, instead of being $\mathcal{O}\left(h^{j+1}\right)$, as might be expected otherwise, turns out to be $\mathcal{O}\left(h^{j+2}\right)$ since $\int_0^h B_j(h,\zeta)W(0)\,\mathrm{d}\zeta$ vanishes. In the case of $\mu_{e,j,1}(h)$ where we take the even part of the potential W at $\zeta-\frac{h}{2}$, this translates into a gain of two powers of h since the smallest non-vanishing term is

$$\frac12\int_0^h B_j(h,\zeta)W^{(2)}(0)\left(\zeta-\frac{h}{2}\right)^2\mathrm{d}\zeta.$$

Consequently, the term $-2\mathrm{i}\varepsilon\left\langle\partial_x^2\mu_{e,2,1}(h)\right\rangle_2$ is $\mathcal{O}\left(\varepsilon^{5\sigma-1}\right)$ in contrast to the $\mathcal{O}\left(\varepsilon^{4\sigma-1}\right)$ term $-2\mathrm{i}\varepsilon\left\langle\partial_x^2\mu_{2,1}(h)\right\rangle_2$ which appears in (5.12). This gain of power arises from discarding the even part $-2\mathrm{i}\varepsilon\left\langle\partial_x^2\mu_{o,2,1}(h)\right\rangle_2$ from the full term.

Similar analysis shows that $\mathrm{i}\varepsilon^{-1}\Lambda\left[\phi\right]_{e,1,1}(h)$ is also $\mathcal{O}\left(\varepsilon^{5\sigma-1}\right)$ since the integral of ϕ over the triangle vanishes. We may, therefore, discard this term from the expansion $\tilde\Theta_4^\varepsilon$. We note that, after discarding the even terms and the $\mathcal{O}\left(\varepsilon^{5\sigma-1}\right)$ odd terms, $\tilde\Theta_4^\varepsilon$ and $\tilde\Theta_3^\varepsilon$ become identical. This is not entirely surprising since both feature an $\mathcal{O}\left(\varepsilon^{5\sigma-1}\right)$ error.

The full power of this approach becomes evident when working on higher-order expansions such as $\tilde\Theta_5^\varepsilon(h)$,

$$\begin{aligned}\tilde\Theta_5^\varepsilon(h)^o &= \overbrace{\mathrm{i}h\varepsilon\partial_x^2+\mathrm{i}\varepsilon^{-1}\mu_{e,0,0}(h)}^{\mathcal{O}\left(\varepsilon^{\sigma-1}\right)}+\overbrace{2\left\langle\partial_x\mu_{o,1,1}(h)\right\rangle_1}^{\mathcal{O}\left(\varepsilon^{3\sigma-1}\right)}\\ &\quad+\overbrace{\mathrm{i}\varepsilon^{-1}\Lambda\left[\phi\right]_{e,1,1}(h)-2\mathrm{i}\varepsilon\left\langle\partial_x^2\mu_{e,2,1}(h)\right\rangle_2}^{\mathcal{O}\left(\varepsilon^{5\sigma-1}\right)}\\ &\quad+\overbrace{\tfrac16\left\langle\Lambda\left[\psi_1+\theta_1\right]_{o,1,2}(h)+\Lambda\left[\psi_2+\theta_2\right]_{o,2,1}(h)\right\rangle_1}^{\mathcal{O}\left(\varepsilon^{5\sigma-1}\right)}\\ &\quad-\overbrace{\tfrac43\varepsilon^2\left\langle\partial_x^3\mu_{o,1,3}(h)\right\rangle_3}^{\mathcal{O}\left(\varepsilon^{5\sigma-1}\right)}-\overbrace{\tfrac14\mathrm{i}\varepsilon\partial_x^4\mu_{e,2,1}(h)}^{\mathcal{O}\left(\varepsilon^{7\sigma-1}\right)}=\Theta(h)+\mathcal{O}\left(\varepsilon^{7\sigma-1}\right).\end{aligned}$$

Since $\psi_j + \theta_j$ is even, $\Big(\Lambda\left[\psi_j + \theta_j\right]_{a,b}(h)\Big)^o = \Lambda\left[\psi_j + \theta_j\right]_{o,a,b}(h)$. We also note that $\Lambda\left[f\right]_{o,a,b}(h)$ and $\mu_{o,j,k}(h)$ gain an extra power of h since the odd part of the function V does not feature a constant term, $V^o(0) = 0$. The term $\mathrm{i}\varepsilon\partial_x^4\mu_{e,2,1}(h)$, which is $\mathcal{O}\left(\varepsilon^{7\sigma-1}\right)$, can now be discarded. In this way, we are able to eradicate all $\mathcal{O}\left(\varepsilon^{4\sigma-1}\right)$ and $\mathcal{O}\left(\varepsilon^{6\sigma-1}\right)$ terms, which correspond to even powers of h. This process allows us to use half the number of terms for the same degree of accuracy.

A Zassenhaus splitting commencing from this Magnus expansion naturally yields an exponential splitting featuring only $\mathcal{O}\left(\varepsilon^{\sigma-1}\right)$, $\mathcal{O}\left(\varepsilon^{3\sigma-1}\right)$ and $\mathcal{O}\left(\varepsilon^{5\sigma-1}\right)$ terms,

$$\exp\left(\tilde{\Theta}_5^{\varepsilon}(h)\right) = \mathrm{e}^{\frac{1}{2}W^{[0]}}\mathrm{e}^{\frac{1}{2}W^{[1]}}\mathrm{e}^{\frac{1}{2}W^{[2]}}\mathrm{e}^{\mathcal{W}^{[3]}}\mathrm{e}^{\frac{1}{2}W^{[2]}}\mathrm{e}^{\frac{1}{2}W^{[1]}}\mathrm{e}^{\frac{1}{2}W^{[0]}} + \mathcal{O}\left(\varepsilon^{7\sigma-1}\right), \qquad (7.12)$$

where

$$\begin{aligned}
W^{[0]} &= \mathrm{i}h\varepsilon\partial_x^2 = \mathcal{O}\left(\varepsilon^{\sigma-1}\right),\\
W^{[1]} &= \mathrm{i}\varepsilon^{-1}\mu_{e,0,0}(h) = \mathcal{O}\left(\varepsilon^{\sigma-1}\right),\\
W^{[2]} &= \tfrac{1}{6}\mathrm{i}h\varepsilon^{-1}\left(\partial_x\mu_{e,0,0}(h)\right)^2 + 2\left\langle\partial_x\mu_{o,1,1}(h)\right\rangle_1 - \tfrac{1}{6}\mathrm{i}h^2\varepsilon\left\langle\partial_x^2\mu_{e,0,0}(h)\right\rangle_2 = \mathcal{O}\left(\varepsilon^{3\sigma-1}\right),\\
\mathcal{W}^{[3]} &= \mathrm{i}\varepsilon^{-1}\Lambda\left[\phi\right]_{e,1,1}(h) + \tfrac{1}{24}\mathrm{i}h^2\varepsilon\left(\partial_x^4\mu_{e,0,0}(h)\right) + \tfrac{1}{6}\mathrm{i}\varepsilon^{-1}\left(\partial_x\mu_{e,0,0}(h)\right)^2\left(\partial_x^2\mu_{e,1,2}(h)\right)\\
&\quad + \tfrac{2}{45}\mathrm{i}h^2\varepsilon^{-1}\left(\partial_x\mu_{e,0,0}(h)\right)^2\left(\partial_x^2\mu_{e,0,0}(h)\right)\\
&\quad + \tfrac{1}{6}\left\langle\Lambda\left[\psi_1+\theta_1\right]_{o,1,2}(h) + \Lambda\left[\psi_2+\theta_2\right]_{o,2,1}(h)\right\rangle_1\\
&\quad + h\left\langle\left(\partial_x\mu_{e,0,0}(h)\right)\left(\partial_x^2\mu_{o,1,1}(h)\right) - \tfrac{1}{3}\left(\partial_x^2\mu_{e,0,0}(h)\right)\left(\partial_x\mu_{o,1,1}(h)\right)\right\rangle_1\\
&\quad + \tfrac{1}{30}\mathrm{i}h^3\varepsilon\left\langle\left(\partial_x^2\mu_{e,0,0}(h)\right)^2 - 2\left(\partial_x\mu_{e,0,0}(h)\right)\left(\partial_x^3\mu_{e,0,0}(h)\right)\right\rangle_2\\
&\quad - 2\mathrm{i}\varepsilon\left\langle\partial_x^2\mu_{e,2,1}(h)\right\rangle_2 - \tfrac{4}{3}\varepsilon^2\left\langle\partial_x^3\mu_{o,3,1}(h)\right\rangle_3\\
&\quad - \tfrac{1}{3}h^2\varepsilon^2\left\langle\partial_x^3\mu_{o,1,1}(h)\right\rangle_3 + \tfrac{1}{120}\mathrm{i}h^4\varepsilon^3\left\langle\partial_x^4\mu_{e,0,0}(h)\right\rangle_4 = \mathcal{O}\left(\varepsilon^{5\sigma-1}\right).
\end{aligned}$$

The term $\frac{1}{24}\mathrm{i}h^2\varepsilon\left(\partial_x^4\mu_{e,0,0}(h)\right)$ in $\mathcal{W}^{[3]}$ is $\mathcal{O}\left(\varepsilon^{3\sigma+1}\right)$. We remind the reader that this splitting is obtained subject to $1/3 < \sigma \leq 1$. For $1/2 < \sigma \leq 1$, this term can be bundled with the $\mathcal{O}\left(\varepsilon^{5\sigma-1}\right)$ terms in $\mathcal{W}^{[3]}$, whereas for $\sigma \leq 1/2$ it can be ignored since it is smaller than $\mathcal{O}\left(\varepsilon^{7\sigma-1}\right)$.

This scheme requires the evaluation of five line integrals,

$$\begin{aligned}
\mu_{e,0,0}(h) &= \int_0^h W^e\left(\zeta - \frac{h}{2}\right)\mathrm{d}\zeta,\\
\mu_{o,1,1}(h) &= \int_0^h \left(\zeta - \tfrac{h}{2}\right)W^o\left(\zeta - \frac{h}{2}\right)\mathrm{d}\zeta,\\
\mu_{e,1,2}(h) &= \int_0^h \left(\zeta - \tfrac{h}{2}\right)^2 W^e\left(\zeta - \frac{h}{2}\right)\mathrm{d}\zeta,\\
\mu_{e,2,1}(h) &= \int_0^h \left(\zeta^2 - h\zeta + \tfrac{1}{6}h^2\right)W^e\left(\zeta - \frac{h}{2}\right)\mathrm{d}\zeta,\\
\mu_{o,3,1}(h) &= \int_0^h \left(\zeta^3 - \tfrac{3}{2}h\zeta^2 + \tfrac{1}{2}h^2\zeta\right)W^o\left(\zeta - \frac{h}{2}\right)\mathrm{d}\zeta,
\end{aligned}$$

and three integrals over the triangle,

$$\int_0^h \int_0^\zeta \phi(h,\zeta,\xi) \left[\partial_x W\left(\zeta-\frac{h}{2}\right)\ \partial_x W\left(\xi-\frac{h}{2}\right)\right]^e \mathrm{d}\xi\,\mathrm{d}\zeta,$$
$$\int_0^h \int_0^\zeta (\psi_1+\theta_1)(h,\zeta,\xi) \left[\partial_x W\left(\zeta-\frac{h}{2}\right)\ \partial_x^2 W\left(\xi-\frac{h}{2}\right)\right]^o \mathrm{d}\xi\,\mathrm{d}\zeta,$$
$$\int_0^h \int_0^\zeta (\psi_2+\theta_2)(h,\zeta,\xi) \left[\partial_x^2 W\left(\zeta-\frac{h}{2}\right)\ \partial_x W\left(\xi-\frac{h}{2}\right)\right]^o \mathrm{d}\xi\,\mathrm{d}\zeta,$$

once per time step. If analytic expressions are not available, it is possible to approximate these through Gauss–Legendre quadrature. For the $\mathcal{O}\left(\varepsilon^{7\sigma-1}\right)$ splitting here, we require merely three Gauss–Legendre knots $t_k = h(1+k\sqrt{3/5})/2, k=-1,0,1$, with weights $w_k = \frac{5}{18}h, \frac{4}{9}h, \frac{5}{18}h$ (Davis & Rabinowitz 1984), respectively, for the $\mathcal{O}\left(h^7\right)$ accuracy which is required of the quadrature.

Evaluation for the integrals over the line, $\mu_{\star,j,k}(h)$, follows directly using these quadrature knots. Integrals over the triangle are also easily evaluated using the same three knots by substituting W with its interpolating polynomial.

Under the scaling $\sigma = 1$, the accuracy of the scheme is $\mathcal{O}\left(\varepsilon^6\right)$ and we require three and two Lanczos iterations, respectively, for exponentiating $W^{[2]}$ and $\mathcal{W}^{[3]}$. As expected, the global L_∞ error is seen to be $\mathcal{O}\left(\varepsilon^{9/2}\right)$ (Figure 6.3) through numerical experiments. The same set-up as Section 6.4 is used for the numerical experiments.

8 Conclusions

In this paper we have presented an effective method for the computation of the Schrödinger equation featuring time-dependent potentials (1.1) under the semiclassical scaling where $\varepsilon \ll 1$. It has led to asymptotic Zassenhaus splittings (6.5) and (7.12), where the exponents are progressively smaller and retain integrals appearing in the Magnus expansion in the undiscretised form. This allows us a great degree of flexibility when handling highly oscillatory potentials. As a consequence of the Lie-algebraic framework introduced here, each argument of an exponential is skew-Hermitian and commutator free. Consequently, the exponentials are unitary and inexpensive to evaluate.

The approach presented in this work should prove general enough to extend to a range of other equations in the Schrödinger family. Some of the more immediate possibilities of extensions of our work are

1. *Semi-discretisation strategies*: while this work has been presented with spectral collocation as the underlying spatial discretisation, the results should prove to be general enough to allow other strategies which suitably take care of the high oscillations of the wavefunction.

2. *Hagedorn wave-packets approach*: to some extent the Hagedorn wave-packets approach (Gradinaru & Hagedorn 2014, Lasser & Troppmann 2014) falls under the umbrella of semi-discretisation strategies. However, it is unique and promising enough to merit a special mention. The specific form of these wave-packets allows techniques of manipulation, exploiting which could require a re-think.

3. *Highly oscillatory potentials*: this work sets the basis for future research on highly oscillatory potentials by demonstrating how integrals can be kept intact through an effective asymptotic splitting. There remains, however, possibility of exploiting the special nature of certain highly oscillatory potentials to a greater advantage.

4. *Stochastic potentials*: stochastic Schrödinger equations arise in the study of quantum trajectories and continuous measurement (Barchielli & Gregoratti 2009), and their numerical solutions have been studied for a variety of cases (Mora 2005, Liu 2013). The stochastic Magnus expansion, which possesses a favourable numerical behaviour (Lord, Malham & Wiese 2008), could allow us to extend our techniques to potentials with stochastic components of certain forms.

References

Abramowitz, M. & Stegun, I. A. (1964), *Handbook of Mathematical Functions with Formulas, Graphs, and Mathematical Tables*, ninth Dover printing, 10th GPO printing edn, Dover, New York.

Bader, P., Iserles, A., Kropielnicka, K. & Singh, P. (2014), 'Effective approximation for the semiclassical Schrödinger equation', *Foundations of Computational Mathematics* **14**, 689–720.

Bao, W., Jin, S. & P. A. Markowich (2002), 'On time-splitting spectral approximations for the Schrödinger equation in the semiclassical regime', *J. Comput. Phys.* **175**, 487–524.

Barchielli, A. & Gregoratti, M. (2009), *Quantum trajectories and measurements in continuous time. The diffusive case*, Vol. 782 of *Lecture Notes in Physics.*, Springer, Heidelberg.

Blanes, S., Casas, F. & Murua, A. (2006), 'Symplectic splitting operator methods tailored for the time-dependent Schrödinger equation', *J. Chem. Phys.* **124**, 105–234.

Blanes, S., Casas, F., Oteo, J. A. & Ros, J. (2009), 'The Magnus expansion and some of its applications', *Phys. Rep.* **470**, 151–238.

Briggs, J. S. & Rost, J. M. (2001), 'On the derivation of the time-dependent equation of Schrödinger', *Foundations of Physics* **31**(4), 693–712.

Casas, F. & Murua, A. (2009), 'An efficient algorithm for computing the Baker–Campbell–Hausdorff series and some of its applications', *J. Math. Phys.* **50**, (electronic).

Davis, P. J. & Rabinowitz, P. (1984), *Methods of Numerical Integration*, 2nd edn, Academic Press, Orlando, FL.

Faou, E. (2012), *Geometric Numerical Integration and Schrödinger Equations*, Zurich Lectures in Advanced Mathematics, Europ. Math. Soc., Zürich.

Fornberg, B. (1998), *A Practical Guide to Pseudospectral Methods*, Cambridge University Press, Cambridge.

Gallopoulos, E. & Saad, Y. (1992), 'Efficient solution of parabolic equations by Krylov approximation methods', *SIAM J. Sci. Statist. Comput* **13**, 1236–1264.

Golub, G. H. & Van Loan, C. F. (1996), *Matrix Computations*, 3rd edn, Johns Hopkins University Press, Baltimore.

Gradinaru, V. & Hagedorn, G. (2014), 'Convergence of a semiclassical wavepacket based time-splitting for the Schrödinger equation', *Numerische Mathematik* **126**(1), 53–73.

Hesthaven, J. S., Gottlieb, S. & Gottlieb, D. (2007), *Spectral Methods for Time-Dependent Problems*, Cambridge University Press, Cambridge.

Hinch, E. J. (1991), *Perturbation Methods*, Cambridge University Press, Cambridge.

Hochbruck, M. & Lubich, C. (1997), 'On Krylov subspace approximations to the matrix exponential operator', *SIAM J. Numer. Anal.* **34**, 1911–1925.

Hochbruck, M. & Lubich, C. (2002), 'On Magnus integrators for time-dependent Schrödinger equations', *SIAM J. Numer. Anal* **41**, 945–963.

Iserles, A. & Nørsett, S. P. (1999), 'On the solution of linear differential equations in Lie groups', *Phil. Trans. Royal Soc. A* **357**, 983–1020.

Iserles, A. & Nørsett, S. P. (2005), 'Efficient quadrature of highly oscillatory integrals using derivatives', *Proceedings of the Royal Society of London A: Mathematical, Physical and Engineering Sciences* **461**(2057), 1383–1399.

Iserles, A., Munthe-Kaas, H. Z., Nørsett, S. P. & Zanna, A. (2000), 'Lie-group methods', *Acta Numerica* **9**, 215–365.

Iserles, A., Nørsett, S. P. & Rasmussen, A. (2001), 'Time symmetry and high-order Magnus methods', *Appl. Numer. Math.* **39**(3–4), 379–401.

Jin, S., Markowich, P. & Sparber, C. (2011), 'Mathematical and computational methods for semiclassical Schrödinger equations', *Acta Numerica* **20**, 121–210.

Lasser, C. & Troppmann, S. (2014), 'Hagedorn wavepackets in time-frequency and phase space', *Journal of Fourier Analysis and Applications* **20**(4), 679–714.

Lehmer, D. H. (1988), 'A new approach to Bernoulli polynomials', *Am. Math. Monthly* **95**(10), 905–911.

Liu, J. (2013), 'A mass-preserving splitting scheme for the stochastic Schrödinger equation with multiplicative noise', *IMA Journal of Numerical Analysis*.

Lord, G., Malham, S. J. A. & Wiese, A. (2008), 'Efficient strong integrators for linear stochastic systems', *SIAM Journal on Numerical Analysis* **46**(6), 2892–2919.

Lubich, C. (2008), *From Quantum to Classical Molecular Dynamics: Reduced Models and Numerical Analysis*, Zurich Lectures in Advanced Mathematics, Europ. Math. Soc., Zürich.

Magnus, W. (1954), 'On the exponential solution of differential equations for a linear operator', *Comm. Pure Appl. Math.* **7**, 649–673.

McLachlan, R. I. & Quispel, G. R. W. (2002), 'Splitting methods', *Acta Numerica* **11**, 341–434.

Moan, P. C. & Niesen, J. (2008), 'Convergence of the Magnus series', *Found. Comput. Math.* **8**(3), 291–301.

Mora, C. M. (2005), 'Numerical solution of conservative finite-dimensional stochastic Schrödinger equations', *Annals of Applied Probability* **15**, 2144–2171.

Munthe–Kaas, H. & Owren, B. (1999), 'Computations in a free Lie algebra', *"Phil. Trans. Royal Soc. A"* **357**(1754), 957–981.

Murua, A. (2010), 'The symmetric Baker–Campbell–Hausdorff formula up to terms of degree 20 written in a Hall basis or a Lyndon basis', `http://www.ehu.es/ccwmuura/research/sbchHall19.dat`.

Oteo, J. A. (1991), 'The Baker–Campbell–Hausdorff formula and nested commutator identities', *J. Math. Phys.* **32**, 419–424.

Reutenauer, C. (1993), *Free Lie Algebras*, London Maths Soc. Monographs **7**, Oxford University Press, Oxford.

Shapiro, M. & Brumer, P. (2003), *Principles of the Quantum Control of Molecular Processes*, Wiley-Interscience, Hoboken, N.J.

Tal Ezer, H. & Kosloff, R. (1984), 'An accurate and efficient scheme for propagating the time dependent Schrödinger equation', *J. Chem. Phys.* **81**, 3967–3976.

Yošida, H. (1990), 'Construction of higher order symplectic integrators', *Phys. Lett.* **150**, 262–268.