Efficient methods for time-dependence in semiclassical Schrödinger equations

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Abstract

We build an efficient and unitary (hence stable) method for the solution of the semiclassical Schrödinger equation subject to explicitly time-dependent potentials. The method is based on a combination of the Zassenhaus decomposition (Bader, Iserles, Kropielnicka & Singh 2014) with the Magnus expansion of the time-dependent Hamiltonian. Our schemes commence from a Magnus expansion where nested integrals are discretised using Gauss–Legendre quadratures following the approach of Munthe-Kaas & Owren (1999). Unlike traditional schemes which typically commence from spatial discretisation, however, our methods are devised by working directly in the free Lie algebra of the undiscretised kinetic and potential operators. Due to the property of height reduction in these Lie algebras, we are able to devise schemes whose costs grow quadratically as we seek higher-order accuracies. These methods also converge despite time-steps being much larger than conventionally considered possible for the convergence of the Magnus expansion.

1 Introduction

Rapid advances in laser technologies over the recent years have led to a significant progress in the control of systems at the molecular level (Shapiro & Brumer 2003). To analyse the control exerted by these lasers we need efficient means of computing the Schrödinger equation featuring time-dependent Hamiltonians (Kormann, Holmgren & Karlsson 2008), existing strategies for which are either low accuracy or become prohibitively expensive with higher orders of accuracy.

We are interested in the numerical computation of the linear, time-dependent Schrödinger equation in a semiclassical regime for a nucleus moving in a time-dependent electric field,

$$\partial_t u(x,t) = -\frac{\mathrm{i}}{\varepsilon} H u(x,t) = \mathrm{i}[\varepsilon\Delta - \varepsilon^{-1}V(x,t)]u(x,t),\ x \in [-1,1],\ t \geq 0, \tag{1.1}$$

equipped with an initial condition $u(x,t) = u_0(x)$, and periodic boundary conditions. We assume, that the potential $V \in C^\infty[-1,1]$ is periodic.

The equation (1.1) is posed on a Hilbert space $\mathcal{H} = \mathrm{L}_2[-1,1]$, and the squared modulus of the solution is the probability density of finding the particle in state x at time t. For this

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reason, the initial density is normalised to one and it is easy to see that the norm of the solution is an invariant,

$$\|u(x,t)\|_{\mathrm{L}^2}^2 = \int_{-1}^{1} |u(x,t)|^2 dx = \|u(x,0)\|_{\mathrm{L}^2}^2.$$

The wave function undergoes unitary evolution, which we wish to preserve under discretisation – both because of physical significance, and since, as we mention in section 4, it implies stability of the numerical method.

Here the parameter $0 < \varepsilon \ll 1$ is the square root of the ratio of masses of the electron and the nucleus. The regularity of V depends on the order of desired accuracy, but for convenience we have assumed that it is smooth in its domain. The initial condition is usually a high-frequency wave packet, but even if it is non-oscillatory it can be shown, cf. the analysis in (Jin, Markowich & Sparber 2011), that the solution to this Schrödinger equation is highly oscillatory, with frequency of at least $\mathcal{O}(\varepsilon^{-1})$. This, as a matter of fact, is the main reason why finding an effective numerical method for (1.1) is such a challenging task. Obviously, the naive approach of finite differences is out of consideration, but instead the usual methodology consists of a semidiscretisation in space followed by an exponential splitting.

Powerful tools like Zassenhaus splitting or Baker–Campbell–Hausdorff formula were historically avoided in splitting methods due to the large computational cost of nested commutators. However as it happens, choosing the correct, infinite–dimensional Lie algebra in case of the Schrödinger vector field, these commutators lose their unwelcome features and enable the derivation of effective, asymptotic splittings.

In (Bader et al. 2014), the current authors established a new framework for a numerical approach to the linear time-dependent problem with an autonomous potential,

$$\partial_t u(x,t) = \mathrm{i}[\varepsilon\Delta - \varepsilon^{-1}V(x)]u(x,t),\ x \in [-1,1],\ t \geq 0,$$

where the underlying problem is considered to evolve in a certain Lie group, and the splitting of the linear operator on the right is followed by semidiscretisation. Due to the choice of a suitable Lie algebra, the authors derived exponential splittings of the form

$$\mathrm{e}^{\mathrm{i}h(\varepsilon\mathcal{K}-\varepsilon^{-1}\mathcal{D})} = \mathrm{e}^{\frac{1}{2}W^{[0]}}\mathrm{e}^{\frac{1}{2}W^{[1]}}\cdots\mathrm{e}^{\frac{1}{2}W^{[s]}}\mathrm{e}^{\mathcal{W}^{[s+1]}}\mathrm{e}^{\frac{1}{2}W^{[s]}}\cdots\mathrm{e}^{\frac{1}{2}W^{[1]}}\mathrm{e}^{\frac{1}{2}W^{[0]}} + \mathcal{O}\left(\varepsilon^{2s+2}\right), \qquad (1.2)$$

where

$$\begin{aligned} W^{[0]} &= W^{[0]}(h,\varepsilon,\mathcal{K},\mathcal{D}) = \mathcal{O}\left(\varepsilon^{0}\right), \\ W^{[k]} &= W^{[k]}(h,\varepsilon,\mathcal{K},\mathcal{D}) = \mathcal{O}\left(\varepsilon^{2k-2}\right), \qquad k = 1,\ldots,s, \\ \mathcal{W}^{[k+1]} &= \mathcal{W}^{[k+1]}(h,\varepsilon,\mathcal{K},\mathcal{D}) = \mathcal{O}\left(\varepsilon^{2s}\right) \end{aligned}$$

and variations on this theme. Here $\mathcal{K}$ and $\mathcal{D}$ are matrices that approximate second differentiation and multiplication by the potential V, respectively. Splittings of the form (1.2) are superior to standard exponential splittings in a number of ways.

First of all, instead of quantifying the errors in terms of the step size, h, which could have been misleading due to large hidden constants, the errors are quantified in terms of the inherent semiclassical parameter ε, taking into account the $\mathcal{O}\left(\varepsilon^{-1}\right)$ oscillations characteristic of the semiclassical Schrödinger equation.

Secondly, these require far fewer exponentials than classical splittings to attain a given order. To be precise, the number of exponentials is shown to grow linearly, rather than

exponentially, with the order. Moreover, the exponents decay increasingly more rapidly in powers of ε, yielding an *asymptotic splitting*.

Thirdly, each of these exponentials can be computed fairly easily. The exponents $W^{[0]}$ and $W^{[1]}$ are either diagonal or circulant matrices and their exponentials can be computed either directly or through FFT, respectively. Remaining exponents are very small and their exponentials can be computed cheaply using low-dimensional Lanczos methods.

The overall cost is quadratic in the desired order, in contrast to the exponential costs of Yošida type splittings which becomes increasingly prohibitive once the Hamiltonian to be split features more than two terms.

To develop such asymptotic splittings for Schrödinger equations with time-varying potentials, we must first resort to the Magnus expansion. We follow the approach of (Munthe-Kaas & Owren 1999) in section 3, discretising the integrals in the Magnus expansion using Gauss-Legendre quadratures. To arrive at a commutator-free expression, however, we must work in the *free Lie algebra* of the infinite dimensional operators ∂_x^2 and V discussed in section 2. Following the framework of (Bader et al. 2014), a Zassenhaus splitting is carried out on the commutator free Magnus expansion. Implementation and numerical examples are discussed in section 4.

An alternative approach was developed in a recent work (Iserles, Kropielnicka & Singh 2015), where the integrals appearing in the Magnus expansion are discretised at the very last stage, following a Zassenhaus splitting.

2 Lie-group setting

Following the established framework in (Bader et al. 2014), we suppress the dependence on x in (1.1) and analyse the following abstract ODE

$$\partial_t u(t) = \mathcal{A}(t)u(t),\ u(0) = u_0, \tag{2.3}$$

where $\mathcal{A}(t) := \mathrm{i}\varepsilon\partial_x^2 - \mathrm{i}\varepsilon^{-1}V(t)$. Because 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 unitary and resides in $\mathcal{U}(\mathcal{H})$ – the Lie group corresponding to $\mathfrak{u}(\mathcal{H})$.

The vector field in the semiclassical Schrödinger equation is a linear combination of the action of two operators, ∂_x^2 and multiplication by the interaction potential V. Since our main tools, Magnus expansion and exponential splitting methods, entail nested commutation, we consider the free Lie algebra,

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

i.e., the linear-space closure of all nested commutators generated by ∂_x^2 and V. Following (Bader et al. 2014), we describe their action on sufficiently smooth functions, e.g.

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

which means that $[V, \partial_x^2] = -(\partial_x^2 V) - 2(\partial_x V)\partial_x$. In general, we note that all terms in $\mathfrak{F}$ belong to the set

$$\mathfrak{G} = \left\{ \sum_{k=0}^{n} y_k(x)\partial_x^k \ :\ n \in \mathbb{Z}_+,\ y_0, \ldots, y_n \in \mathrm{C}_p^\infty[-1,1] \right\},$$

where the subscript p means periodicity in $[-1, 1]$. It is trivial to observe that $\mathfrak{G}$ is itself a

Lie algebra with the commutator

$$\left[\sum_{i=0}^{n} f_i(x)\partial_x^i, \sum_{j=0}^{m} g_j(x)\partial_x^j\right] = \sum_{i=0}^{n}\sum_{j=0}^{m}\sum_{\ell=0}^{i}\binom{i}{\ell} f_i(x)\left(\partial_x^{i-\ell} g_j(x)\right)\partial_x^{\ell+j} - \sum_{j=0}^{m}\sum_{i=0}^{n}\sum_{\ell=0}^{j}\binom{j}{\ell} g_j(x)\left(\partial_x^{j-\ell} f_i(x)\right)\partial_x^{\ell+i}. \tag{2.4}$$

In similar vein to (Bader et al. 2014), we proceed in the pursuit of stability to replace all odd powers of ∂_x that are accompanied by i. The identities,

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

where y is a C^1 function, suffice for our presentation. The general form for expressing $y\partial_x^{2s+1}$ as a linear combination of even derivatives is reported in (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 our operators ∂_x^2 and V are each multiplied 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 is not enough to blindly 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 the replacement of even derivatives by odd derivatives can be proven along similar lines as (Bader et al. 2014). For all practical purposes, however, we only require the identities

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

which can be easily verified directly.

Once 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, they are 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 convenient 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). 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 will feature in another publication, but not much is lost here by considering these as merely a notational convenience. For the purpose of this work we make observations which can be verified using the machinery of (2.4) in conjunction with the odd and even derivative replacement rules. We present identities which suffice for simplifying all commutators appearing in this work,

$$\begin{aligned}
&[\langle f\rangle_4, \langle g\rangle_0] = 4\langle f(\partial_x g)\rangle_3 - 2\left\langle 3(\partial_x f)(\partial_x^2 g) + f(\partial_x^3 g)\right\rangle_1, \\
&[\langle f\rangle_3, \langle g\rangle_0] = 3\langle f(\partial_x g)\rangle_2 - \tfrac{1}{2}\left\langle 3(\partial_x f)(\partial_x^2 g) + f(\partial_x^3 g)\right\rangle_0, \\
&[\langle f\rangle_2, \langle g\rangle_2] = 2\langle f(\partial_x g) - (\partial_x f)g\rangle_3 + \left\langle 2(\partial_x^2 f)(\partial_x g) - 2(\partial_x f)(\partial_x^2 g) + (\partial_x^3 f)g - f(\partial_x^3 g)\right\rangle_1, \\
&[\langle f\rangle_2, \langle g\rangle_1] = \langle 2f(\partial_x g) - (\partial_x f)g\rangle_2 - \tfrac{1}{2}\left\langle 2(\partial_x f)(\partial_x^2 g) + f(\partial_x^3 g)\right\rangle_0, \\
&[\langle f\rangle_2, \langle g\rangle_0] = 2\langle f(\partial_x g)\rangle_1, \\
&[\langle f\rangle_1, \langle g\rangle_1] = \langle f(\partial_x g) - (\partial_x f)g\rangle_1, \\
&[\langle f\rangle_1, \langle g\rangle_0] = \langle f(\partial_x g)\rangle_0.
\end{aligned} \tag{2.5}$$

The terms $\mathrm{i}\partial_x^2 = \mathrm{i}\langle 1\rangle_2$ and $\mathrm{i}V = \mathrm{i}\langle V\rangle_0$ reside in

$$\mathfrak{H} = \{\mathrm{i}^{k+1}\langle f\rangle_k \;:\; f \in \mathrm{C}_p^\infty([-1,1];\mathbb{R}),\ k \geq 0\}$$

and, as evident through a few examples in (2.5), all commutators of elements of $\mathfrak{H}$ also reside in $\mathfrak{H}$. In other words, $\mathfrak{H}$ is a Lie algebra such that

$$\mathrm{FLA}\{\mathrm{i}\partial_x^2, \mathrm{i}V\} \subseteq \mathfrak{H},$$

and it suffices to work directly in $\mathfrak{H}$ using the rules (2.5) instead of proceeding via (2.4) followed by the odd-even derivative replacement rules.

For a real valued f, $\langle f\rangle_k$ is symmetric if k is even and skew-symmetric otherwise. This property is preserved under discretisation once we use spectral collocation on a uniform grid. In that case ∂_x is discretised as a skew-symmetric matrix $\mathcal{K}$ and V is discretised as a diagonal matrix $\mathcal{D}_V$. The term $\langle f\rangle_k$ is discretised as $\left(\mathcal{K}^k\mathcal{D}_f + \mathcal{D}_f\mathcal{K}^k\right)/2$ which is clearly symmetric when k is even and skew-symmetric otherwise. Consequently, elements of $\mathfrak{H}$ such as $\mathrm{i}^{k+1}\langle f\rangle_k$, which are skew-Hermitian operators, discretise to skew-Hermitian matrices of the form $\mathrm{i}^{k+1}\left(\mathcal{K}^k\mathcal{D}_f + \mathcal{D}_f\mathcal{K}^k\right)/2$.

This structural property of $\mathfrak{H}$ is responsible for unitary evolution and numerical stability of our schemes since exponentials of skew-Hermitian matrices are unitary.

Definition 1 *The height of a term is defined as*

$$\mathrm{ht}\left(\sum_{i=0}^{n} \alpha_i \langle f_i\rangle_{k_i}\right) = \max\{k_1, \ldots, k_n\}.$$

These terms benefit from a remarkable property of *height reduction* which is stated here without proof,

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

For the commutators relevant to this work, this property can be verified by a quick inspection of the identities (2.5).

For the largest part, our work will proceed in the language of the undiscretised operators introduced in this section. At the very last stage we will resort to spectral collocation on the uniform grid over $[-1, 1]$ for spatial discretisation. For this purpose we will need at least $M = \mathcal{O}\left(\varepsilon^{-1}\right)$ points since (regardless of initial conditions) the solution of the Schrödinger equation develops spatial oscillations of order $\mathcal{O}\left(\varepsilon^{-1}\right)$ (Jin et al. 2011, Bao, Jin & Markowich 2002). Consequently, $\mathcal{K}$ scales like $\mathcal{O}\left(\varepsilon^{-1}\right)$ and $\left(\mathcal{K}^k\mathcal{D}_f + \mathcal{D}_f\mathcal{K}^k\right)/2 = \mathcal{O}\left(\varepsilon^{-k}\right)$. Keeping eventual discretisation in mind, we abuse notation and write $\langle f\rangle_k = \mathcal{O}\left(\varepsilon^{-k}\right)$.

The property of height reduction leads to a systematic decrease in the size of terms with commutation,

$$\left[\langle f\rangle_k, \langle g\rangle_l\right] = \mathcal{O}\left(\varepsilon^{-k-l+1}\right).$$

Going further, we want to analyse all terms in the common *currency* of the inherent semiclassical parameter ε and assume that our choice of the time-step, h, is governed by $h = \mathcal{O}\left(\varepsilon^{\sigma}\right)$, for some $0 < \sigma \leq 1$. Larger values of σ correspond to very small time steps and are best avoided.

3 The solution

3.1 The Magnus expansion

To look for the solution of (2.3) one needs to take into account some features of the operator $\mathcal{A}(t)$. First of all it depends on time and it cannot be assumed that its values in different points of time commute, i.e. we assume that $[\mathcal{A}(t_1), \mathcal{A}(t_2)] \neq 0$ and give up the hope that the solution is of the simple form $\mathrm{e}^{\int_0^t \mathcal{A}(\xi)d\xi}u_0$. Secondly $\mathcal{A}(t)$ evolves in a Lie algebra so the solution of (2.3) resides in a corresponding Lie group. Both properties can be dealt with elegantly using the famous result from (Magnus 1954) by writing the solution as single exponential,

$$u(t) = \mathrm{e}^{\Theta(t)}u(0), \tag{3.6}$$

where the infinite series $\Theta(t) = \sum_{k=1}^{\infty}\Theta_k(t)$, also called as *Magnus expansion*, is an element of the underlying Lie algebra. Its convergence has been shown in (Iserles & Nørsett 1999), (Moan & Niesen 2008), (Hochbruck & Lubich 2003) for sufficiently small time–steps. Obviously we truncate this series and advance with adequately small time step h

$$u(t+h) = \mathrm{e}^{\Theta(t+h,t)}u(t), \tag{3.7}$$

starting from the initial step,

$$u(h) = \mathrm{e}^{\Theta(h,0)}u(0), \tag{3.8}$$

where we understand that the operator $\mathrm{e}^{\Theta(t+h,t)}$ is a flow evolving the solution from t to $t+h$. For the sake of simplicity we will analyse the Magnus expansion at the first step only, i.e. (3.8), and shorten the notation, writing $\Theta(h)$ instead of $\Theta(h, 0)$.

Simple differentiation of the *ansatz* in (3.8) together with elementary algebra, see (Iserles & Nørsett 1999) or (Blanes, Casas, Oteo & Ros 2009) for details, lead to the conclusion that

the exponent $\Theta(t)$ satisfies the *dexpinv equation*,

$$\dot{\Theta}(h) = \mathrm{dexp}^{-1}_{\Theta(h)}\mathcal{A}(h) = \sum_{k=0}^{\infty} \frac{B_k}{k!}\mathrm{ad}^k_{\Theta(h)}\mathcal{A}(h), \qquad \Theta(0) = 0, \tag{3.9}$$

where B_k are Bernoulli numbers ($B_0 = 1,\ B_1 = -\frac{1}{2},\ B_2 = \frac{1}{6}\ B_3 = 0,\ B_4 = -\frac{1}{30},\ B_5 = 0,\ B_6 = \frac{1}{42}$) and the adjoint representation is defined recursively by $\mathrm{ad}^0_A V = V$ and $\mathrm{ad}^{k+1}_A V = [A, \mathrm{ad}^k_A V]$. The solution of (3.9) is an infinite series and can be obtained using Picard iterations. It was proposed in (Magnus 1954) and widely analysed in (Iserles & Nørsett 1999, Iserles, Munthe-Kaas, Nørsett & Zanna 2000, Blanes et al. 2009).

The first few terms of the Magnus expansion ordered by size in h are

$$\begin{aligned}
\Theta(h) = &\int_0^h \mathcal{A}(\xi)d\xi \\
&- \frac{1}{2}\int_0^h \int_0^{\xi_1} [\mathcal{A}(\xi_2), \mathcal{A}(\xi_1)]d\xi \\
&+ \frac{1}{12}\int_0^h \int_0^{\xi_1} \int_0^{\xi_1} [\mathcal{A}(\xi_2), [\mathcal{A}(\xi_3), \mathcal{A}(\xi_1)]]d\xi \\
&+ \frac{1}{4}\int_0^h \int_0^{\xi_1} \int_0^{\xi_2} [[\mathcal{A}(\xi_3), \mathcal{A}(\xi_2)], \mathcal{A}(\xi_1)]d\xi \\
&- \frac{1}{24}\int_0^h \int_0^{\xi_1} \int_0^{\xi_1} \int_0^{\xi_3} [\mathcal{A}(\xi_2), [[\mathcal{A}(\xi_4), \mathcal{A}(\xi_3)], \mathcal{A}(\xi_1)]]d\xi \\
&- \frac{1}{24}\int_0^h \int_0^{\xi_1} \int_0^{\xi_2} \int_0^{\xi_2} [[\mathcal{A}(\xi_3), [\mathcal{A}(\xi_4), \mathcal{A}(\xi_2)]], \mathcal{A}(\xi_1)]d\xi \\
&- \frac{1}{8}\int_0^h \int_0^{\xi_1} \int_0^{\xi_2} \int_0^{\xi_3} [[[[\mathcal{A}(\xi_4), \mathcal{A}(\xi_3)], \mathcal{A}(\xi_2)], \mathcal{A}(\xi_1)]d\xi \\
&+ \cdots .
\end{aligned} \tag{3.10}$$

We say that a multivariate integral of a nested commutator, $\mathcal{I}$, is of grade m if $\mathcal{I} = \mathcal{O}(h^m)$ for every smooth $\mathcal{A}$. Truncating the Magnus expansion at grade p to $\Omega_p(h) = \Theta(h) + \mathcal{O}(h^{p+1})$, preserves time symmetry (Iserles et al. 2000), (Iserles, Nørsett & Rasmussen 2001). Time symmetry means that not only the exact flow φ, but also the numerical flow $\Phi = \mathrm{e}^{\Omega_p(h)}$, satisfy

$$\varphi(h, 0) \circ \varphi(0, h) = \boldsymbol{I}, \qquad \Phi_{h,0} \circ \Phi_{0,h} = \boldsymbol{I}. \tag{3.11}$$

As one can observe, the time symmetry of the numerical flow is equivalent to the fact that

$$\Omega_p(0, h) = -\Omega_p(h, 0). \tag{3.12}$$

Time symmetry is a desirable feature because truncation by power with odd p leads to a gain of an extra unit of order, see (Iserles et al. 2000). This means that if we aim for a numerical method of order six it suffices to consider the truncation of the Magnus expansion only to the terms listed in (3.10).

3.2 Magnus expansion in practice

It turns out that the multivariate integrals can be efficiently computed using simple univariate quadrature rules of Munthe-Kaas & Owren (1999). We will follow their approach and evaluate

the potential at the Gauss–Legendre quadrature points ($t_1 = \frac{1}{2} - \frac{\sqrt{15}}{10}$, $t_2 = \frac{1}{2}$, $t_3 = \frac{1}{2} + \frac{\sqrt{15}}{10}$) which is then transformed (Iserles & Nørsett 1999) to obtain a far less costly quadrature. As a result, to obtain order six approximation, all the effort of approximation of the solution boils down to the following formula

$$\begin{aligned}\Theta(h) =& B_1 + \frac{1}{12}B_3 - \frac{1}{12}[B_1, B_2] + \frac{1}{240}[B_2, B_3] + \frac{1}{360}[B_1, [B_1, B_3]] \\ &+ \frac{1}{240}[B_2, [B_1, B_2]] + \frac{1}{720}[B_1, [B_1, [B_1, B_2]]] + \mathcal{O}\left(h^7\right),\end{aligned} \tag{3.13}$$

where

$$B_1 = h\mathcal{A}(t_2),\ B_2 = \frac{\sqrt{15}}{3}h(\mathcal{A}(t_3) - \mathcal{A}(t_1)),\ B_3 = \frac{10}{3}h(\mathcal{A}(t_3) - 2\mathcal{A}(t_2) + \mathcal{A}(t_1)). \tag{3.14}$$

See (Iserles et al. 2000) and (Blanes et al. 2009) for comprehensive information and ways to approximate the Magnus expansion using different quadrature rules and to higher orders. The former could be relevant if the time-dependent potential is only known at certain grid-points as might be the case in some control setups.

Substituting $\mathcal{A}(t)$ with the given Hamiltonian as $\mathcal{A}(t) = -\mathrm{i}H(t)/\varepsilon$ and working in the free Lie algebra $\mathfrak{H}$, we can derive a commutator free expansion using the identities (2.5). Keeping the notation of the previous section in mind, we approximate the time derivatives of the potential by central differences, cf. (3.14),

$$V_0 = V(t_2),\ V_1 = \frac{\sqrt{15}}{3h}(V(t_3) - V(t_1)),\ V_2 = \frac{10}{3h^2}(V(t_3) - 2V(t_2) + V(t_1)),$$

so that

$$B_1 = \mathrm{i}h\varepsilon\partial_x^2 - \mathrm{i}h\varepsilon^{-1}V_0, \quad B_2 = \mathrm{i}h^2\varepsilon^{-1}V_1, \quad B_3 = \mathrm{i}h^3\varepsilon^{-1}V_2.$$

Once these are substituted in (3.13), we use the identities (2.5) along with the observation that $\partial_x^2 = \langle 1\rangle_2$ and $V_j = \langle V_j\rangle_0$ to arrive at a Magnus expansion in the format $\sum_k \mathrm{i}^{k+1}c_k\langle f_k\rangle_k$ with $c_k \in \mathbb{Q}$ and $f_k \in \mathrm{C}_p^\infty([-1,1];\mathbb{R})$,

$$\begin{aligned}\Omega_5 = &\overbrace{\mathrm{i}h\varepsilon\partial_x^2 - \mathrm{i}h\varepsilon^{-1}V_0}^{\mathcal{O}(\varepsilon^0)} - \overbrace{\frac{1}{12}\mathrm{i}h^3\varepsilon^{-1}V_2 - \frac{1}{6}h^3\langle\partial_x V_1\rangle_1}^{\mathcal{O}(\varepsilon^2)} \\ &+ \overbrace{\frac{1}{360}\mathrm{i}h^5\varepsilon^{-1}\Big(2(\partial_x V_2)(\partial_x V_0) - 3(\partial_x V_1)^2\Big)}^{\mathcal{O}(\varepsilon^4)} \\ &- \overbrace{\frac{1}{180}h^5\left\langle(\partial_x V_1)(\partial_x^2 V_0) + 3(\partial_x V_0)(\partial_x^2 V_1)\right\rangle_1}^{\mathcal{O}(\varepsilon^4)} \\ &+ \overbrace{\frac{1}{90}\mathrm{i}h^5\varepsilon\left\langle\partial_x^2 V_2\right\rangle_2 - \frac{1}{90}h^5\varepsilon^2\left\langle\partial_x^3 V_1\right\rangle_3}^{\mathcal{O}(\varepsilon^4)} = \Theta + \mathcal{O}\left(\varepsilon^6\right).\end{aligned}$$

Since Ω_5 includes the term $\mathrm{i}h\varepsilon\partial_x^2 - \mathrm{i}h\varepsilon^{-1}V_0$, its exponential is, at the very least, as troublesome to approximate as the problem of solving the Schrödinger equation with time-independent potential. Fortunately the Zassenhaus procedure is sufficiently flexible and can tackle such modified Hamiltonians with ease.

3.3 Zassenhaus

Let us recall the basic principle for the iterative symmetric Zassenhaus splitting (Bader et al. 2014). Our goal is to compute $e^{\mathcal{W}^{[0]}}$, where $\mathcal{W}^{[0]} = X + Y$ and $X, Y = \mathcal{O}(\varepsilon^p)$. Using the *symmetric Baker-Campbell-Hausdorff* (sBCH) formula (Dynkin 1947, Casas & Murua 2009), we then write

$$e^{\mathcal{W}^{[0]}} = e^{\frac{1}{2}X} e^{\mathrm{sBCH}(-X, \mathcal{W}^{[0]})} e^{\frac{1}{2}X}. \tag{3.15}$$

Grade-three commutators of X and Y are at most $\mathcal{O}\left(\varepsilon^{3p}\right)$ so that $\mathrm{sBCH}(-X, X+Y) = Y + \mathcal{O}\left(\varepsilon^{3p}\right)$. Thus we have extracted X from the exponent at the cost of correction terms in form of higher-order commutators. Assuming that the corrections are decreasing in size, it is then enough to identify the largest terms as $W^{[1]}$ in the central exponent $\mathcal{W}^{[1]} = \mathrm{sBCH}(-X, \mathcal{W}^{[0]})$ and to continue the iteration until the desired accuracy is reached,

$$\mathcal{W}^{[k+1]} = \mathrm{sBCH}(-W^{[k]}, \mathcal{W}^{[k]}), \quad \mathcal{W}^{[0]} = X + Y. \tag{3.16}$$

In this notation, the splitting after s steps can be written as

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

We emphasize that, in principle, we can freely choose the elements $W^{[k]}$ that we want to extract. Except for some special cases, at least one of the exponents in this splitting will feature an infinite series of terms. To construct a finite splitting scheme featuring a certain accuracy we may discard, at each stage, all terms smaller than the desired threshold.

Assuming that a grade k commutator of X and Y scales as $\mathcal{O}\left(\varepsilon^{kp}\right)$, convergence of the series requires $p > 0$ at the very least. In the case of the Schrödinger equation, we choose $X = \mathrm{i}h\varepsilon\partial_x^2$, $p = \sigma - 1$, and this naively translates to a very stringent time step restriction: $\sigma > 1$. However, the remarkable feature of height reduction means that a grade k commutator in this context scales as $\mathcal{O}\left(\varepsilon^{kp+(k-1)}\right)$ and convergence requirements become significantly milder: we need $p + 1 > 0$ which translates to $\sigma > 0$.

3.4 Zassenhaus on Magnus

We perform a Zassenhaus splitting on Ω_5, choosing to extract the largest terms – analysed in powers of ε – first. We commence the splitting with $W^{[0]} = \mathrm{i}h\varepsilon\partial_x^2$, although we could equally well choose $W^{[0]} = -\mathrm{i}h\varepsilon^{-1}V_0$, for instance, and arrive at a variant of the splitting presented here. The exponent to be split is $\mathcal{W}^{[0]} = \Omega_5$ and the first step involves computing the sBCH formula. Here, once again, the rules of the free Lie algebra $\mathfrak{H}$, (2.5), suffice for arriving at a

commutator free expression,

$$
\begin{aligned}
\mathcal{W}^{[1]} &= \mathrm{sBCH}(-W^{[0]}, \mathcal{W}^{[0]}) \\
&= -\overbrace{\mathrm{i}h\varepsilon^{-1}V_0}^{\mathcal{O}\left(\varepsilon^{\sigma-1}\right)} + \overbrace{\frac{1}{12}\mathrm{i}h^3\varepsilon^{-1}\Big(2(\partial_x V_0)^2 - V_2\Big) - \frac{1}{6}h^3\left\langle \partial_x V_1\right\rangle_1 + \frac{1}{6}\mathrm{i}h^3\varepsilon\left\langle \partial_x^2 V_0\right\rangle_2}^{\mathcal{O}\left(\varepsilon^{3\sigma-1}\right)} \\
&\quad - \overbrace{\frac{1}{24}\mathrm{i}h^3\varepsilon(\partial_x^4 V_0)}^{\mathcal{O}\left(\varepsilon^{3\sigma+1}\right)} - \overbrace{\frac{1}{360}\mathrm{i}h^5\varepsilon^{-1}\Big(8(\partial_x V_0)^2(\partial_x^2 V_0) + 3(\partial_x V_1)^2 - 12(\partial_x V_2)(\partial_x V_0)\Big)}^{\mathcal{O}\left(\varepsilon^{5\sigma-1}\right)} \\
&\quad + \overbrace{\frac{1}{30}h^5\left\langle 2(\partial_x V_0)(\partial_x^2 V_1) - (\partial_x V_1)(\partial_x^2 V_0)\right\rangle_1}^{\mathcal{O}\left(\varepsilon^{5\sigma-1}\right)} \\
&\quad - \overbrace{\frac{1}{720}\mathrm{i}h^5\varepsilon\left\langle 127(\partial_x V_0)(\partial_x^3 V_0) + 130(\partial_x^2 V_0)^2 - 18(\partial_x^2 V_2)\right\rangle_2}^{\mathcal{O}\left(\varepsilon^{5\sigma-1}\right)} \\
&\quad + \overbrace{\frac{1}{60}h^5\varepsilon^2\left\langle \partial_x^3 V_1\right\rangle_3 - \frac{13}{90}\mathrm{i}h^5\varepsilon^3\left\langle \partial_x^4 V_0\right\rangle_4}^{\mathcal{O}\left(\varepsilon^{5\sigma-1}\right)} + \mathcal{O}\left(\varepsilon^{7\sigma-1}\right).
\end{aligned}
$$

At the second stage we select the largest remaining element $W^{[1]} = -\mathrm{i}h\varepsilon^{-1}V_0$, whereby

$$
\begin{aligned}
\mathcal{W}^{[2]} &= \mathrm{sBCH}(-W^{[1]}, \mathcal{W}^{[1]}) \\
&= \overbrace{\frac{1}{12}\mathrm{i}h^3\varepsilon^{-1}\Big(2(\partial_x V_0)^2 - V_2\Big) - \frac{1}{6}h^3\left\langle \partial_x V_1\right\rangle_1 + \frac{1}{6}\mathrm{i}h^3\varepsilon\left\langle \partial_x^2 V_0\right\rangle_2}^{\mathcal{O}\left(\varepsilon^{3\sigma-1}\right)} \\
&\quad - \overbrace{\frac{1}{24}\mathrm{i}h^3\varepsilon(\partial_x^4 V_0)}^{\mathcal{O}\left(\varepsilon^{3\sigma+1}\right)} - \overbrace{\frac{1}{360}\mathrm{i}h^5\varepsilon^{-1}\Big(13(\partial_x V_0)^2(\partial_x^2 V) + 3(\partial_x V_1)^2 - 12(\partial_x V_2)(\partial_x V_0)\Big)}^{\mathcal{O}\left(\varepsilon^{5\sigma-1}\right)} \\
&\quad + \overbrace{\frac{1}{30}h^5\left\langle 2(\partial_x V_0)(\partial_x^2 V_1) - (\partial_x V_1)(\partial_x^2 V_0)\right\rangle_1}^{\mathcal{O}\left(\varepsilon^{5\sigma-1}\right)} \\
&\quad - \overbrace{\frac{1}{720}\mathrm{i}h^5\varepsilon\left\langle 127(\partial_x V_0)(\partial_x^3 V_0) + 130(\partial_x^2 V_0)^2 - 18(\partial_x^2 V_2)\right\rangle_2}^{\mathcal{O}\left(\varepsilon^{5\sigma-1}\right)} \\
&\quad + \overbrace{\frac{1}{60}h^5\varepsilon^2\left\langle \partial_x^3 V_1\right\rangle_3 - \frac{13}{90}\mathrm{i}h^5\varepsilon^3\left\langle \partial_x^4 V_0\right\rangle_4}^{\mathcal{O}\left(\varepsilon^{5\sigma-1}\right)} + \mathcal{O}\left(\varepsilon^{7\sigma-1}\right).
\end{aligned}
$$

We terminate the procedure by letting $W^{[2]}$ consist of the $\mathcal{O}\left(\varepsilon^{3\sigma-1}\right)$ terms in $\mathcal{W}^{[2]}$ and are left with $\mathcal{O}\left(\varepsilon^{5\sigma-1}\right)$ and $\mathcal{O}\left(\varepsilon^{3\sigma+1}\right)$ terms in $\mathcal{W}^{[3]} = \mathcal{W}^{[2]} - W^{[2]}$ once we ignore $\mathcal{O}\left(\varepsilon^{7\sigma-1}\right)$ terms. Since $\mathcal{O}\left(\varepsilon^{3\sigma+1}\right)$ terms can be subsumed into the $\mathcal{O}\left(\varepsilon^{5\sigma-1}\right)$ terms for $\sigma \leq 1$, combining them in this way is not a cause for concern. The outcome is the splitting,

$$
\mathrm{e}^{\Omega_5} = e^{\frac{1}{2}W^{[0]}} e^{\frac{1}{2}W^{[1]}} e^{\frac{1}{2}W^{[2]}} e^{\mathcal{W}^{[3]}} e^{\frac{1}{2}W^{[2]}} e^{\frac{1}{2}W^{[1]}} e^{\frac{1}{2}W^{[0]}} + \mathcal{O}\left(\varepsilon^6\right), \tag{3.17}
$$

with

$$\begin{aligned}
W^{[0]} &= \mathrm{i}\varepsilon h\partial_x^2 = \mathcal{O}\left(\varepsilon^{\sigma-1}\right),\\
W^{[1]} &= -\mathrm{i}\varepsilon^{-1}hV_0 = \mathcal{O}\left(\varepsilon^{\sigma-1}\right),\\
W^{[2]} &= \frac{1}{12}\mathrm{i}\varepsilon^{-1}h^3\Big(2(\partial_xV_0)^2 - V_2\Big) - \frac{1}{6}h^3\left\langle\partial_xV_1\right\rangle_1 + \frac{1}{6}\mathrm{i}\varepsilon h^3\left\langle\partial_x^2V_0\right\rangle_2 = \mathcal{O}\left(\varepsilon^{3\sigma-1}\right),\\
\mathcal{W}^{[3]} &= -\frac{1}{24}\mathrm{i}\varepsilon h^3(\partial_x^4V_0) - \frac{1}{360}\mathrm{i}\varepsilon^{-1}h^5\Big(13(\partial_xV_0)^2(\partial_x^2V_0) + 3(\partial_xV_1)^2 - 12(\partial_xV_2)(\partial_xV_0)\Big)\\
&\quad + \frac{1}{30}h^5\left\langle 2(\partial_xV)(\partial_x^2V_1) - (\partial_xV_1)(\partial_x^2V)\right\rangle_1\\
&\quad - \frac{1}{720}\mathrm{i}\varepsilon h^5\left\langle 127(\partial_xV_0)(\partial_x^3V_0) + 130(\partial_x^2V_0)^2 - 18(\partial_x^2V_2)\right\rangle_2\\
&\quad + \frac{1}{60}\varepsilon^2h^5\left\langle\partial_x^3V_1\right\rangle_3 - \frac{13}{90}\mathrm{i}\varepsilon^3h^5\left\langle\partial_x^4V_0\right\rangle_4 = \mathcal{O}\left(\varepsilon^{5\sigma-1}\right).
\end{aligned}$$

4 A numerical scheme

For numerical realisation it is typical to impose periodic boundary conditions in order to resolve spatial oscillations with spectral accuracy. Recall that we restrict the domain to $[-1,1]$, imposing periodic boundaries at $x=\pm1$. We discretise using spectral collocation. The unknowns are $u_n \approx u(n/(N+\frac{1}{2}))$, $|n|\le N$, where $M=2N+1$. The differential operator ∂_x is discretised as a circulant matrix $\mathcal{K}$ and V as a diagonal $\mathcal{D}_V$.

All exponents in our splitting (3.17) are of the form $\mathrm{i}^{k+1}\left\langle f\right\rangle_k$ and are discretised as skew-Hermitian matrices. Since the exponential of a skew-Hermitian matrix is unitary, unitary evolution and (consequently) unconditional stability of the method are guaranteed.

The outermost exponentials $W^{[0]}$ and $W^{[1]}$ are replaced by the circulant $\mathrm{i}\varepsilon h\mathcal{K}^2$ and the diagonal matrix $-\mathrm{i}\varepsilon^{-1}h\mathcal{D}_{V_0}$, respectively. The exponential of a circulant matrix is evaluated to machine precision using Fast Fourier Transform (FFT) in $\mathcal{O}(M\log M)$ operations while a diagonal matrix is exponentiated directly in $\mathcal{O}(M)$ operations. The remaining exponents, $W^{[2]}$ and $\mathcal{W}^{[3]}$, do not posses a structure amenable to exact exponentiation. However, they are very small – $\mathcal{O}\left(\varepsilon^{3\sigma-1}\right)$ and $\mathcal{O}\left(\varepsilon^{5\sigma-1}\right)$, respectively. For $\sigma=1$, the most costly case we consider, the exponentials of these terms can be evaluated to $\mathcal{O}\left(\varepsilon^6\right)$ accuracy using merely three and two Lanczos iterations, respectively.

We refer the curious reader to (Bader et al. 2014, Iserles et al. 2015) where semi-discretisation strategies, stability analysis and exponentiation methods are addressed in greater detail.

4.1 A numerical example

Consider the evolution of the wave-packet

$$u_0(x) = (\delta\pi)^{-1/4}\exp\left(\mathrm{i}k_0\frac{(x-x_0)}{\delta} - \frac{(x-x_0)^2}{2\delta}\right)$$

with $x_0=-0.3$, $k_0=0.1$ and $\delta=1.22\times10^{-4}$, heading towards the lattice potential

$$V_0 = \rho(4x)\sin(20\pi x),$$

where

$$\rho(x) = \begin{cases}\exp\left(-1/(1-x^2)\right) & \text{for } |x|<1,\\ 0 & \text{otherwise,}\end{cases}$$

is a bump function. When the semiclassical parameter is $\varepsilon = 2^{-8}$, the wave-packet evolves to $u(T)$ at $T = 0.75$ (Figure 4.1) under the influence of the time-independent potential V_0 alone. When we excite it using an additional time-varying potential,

$$E(x,t) = \rho(3t-1)\rho(\sin(2\pi(x-t))),$$

so that the wave packet evolves under $V_E(x,t) = V_0(x) + E(x,t)$, a significantly larger part of the wave packet is able to make it across the lattice to the right hand side (see $u_E(T)$ in Figure 4.1).

The excitation pulse is not active for the entire duration since $\rho(3t-1)$ acts as a smooth envelope simulating the switching on and off of the time-varying component of the potential. The excited potential is evident at $t = T/2$ in Figure 4.2.

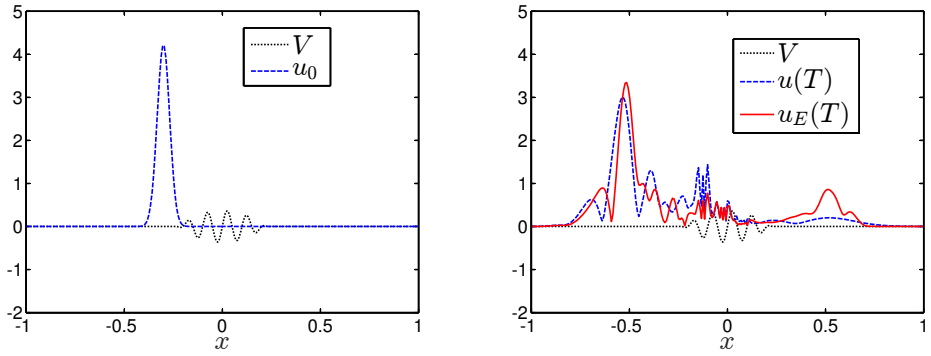

Figure 4.1: (left) initial wave-packet u_0; (right) final wave-packets at time $T = 0.75$: $u(T)$ under the influence of V_0 and $u_E(T)$ under the influence of $V_E(x,t) = V_0 + E(x,t)$.

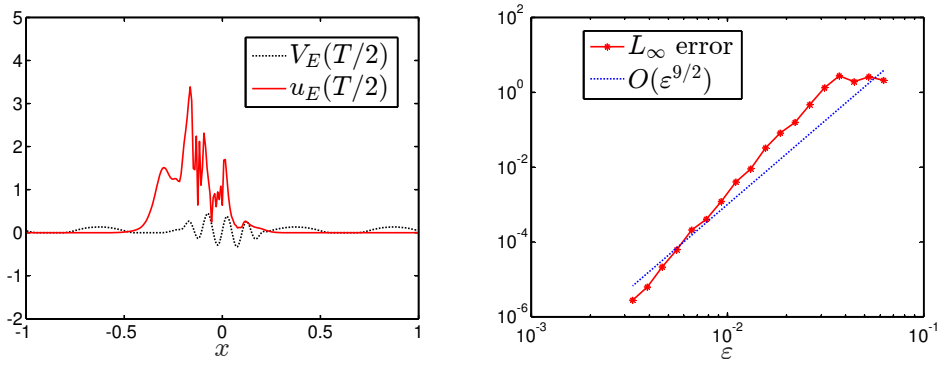

Figure 4.2: (left) effective potential at the middle of the time interval, $t = T/2$ (where $T = 0.75$); (right) global error of the Zassenhaus splitting (3.17) at $T = 0.5$.

In Figure 4.2 we present the global error at time $T = 0.5$ in the propagation of u_0 to u_E under the influence of V_E using the scheme (3.17). Under the scaling $\sigma = 1$, we commit a local L_2 error of $\mathcal{O}\left(\varepsilon^4\right)$ per time step in the splitting scheme (3.17). Since the number of time steps is $\mathcal{O}\left(\varepsilon^{-\sigma}\right)$, the global error is $\mathcal{O}\left(\varepsilon^5\right)$. The precise scaling used in our experiments is $M \sim 5\varepsilon^{-1}$ and $h \sim 2\varepsilon$.

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 $\mathcal{O}\left(\varepsilon^{9/2}\right)$ for $\sigma = 1$. This is indeed seen to be the case through numerical experiments in Figure 4.2.

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