A time-splitting for the semiclassical Schrödinger equation

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Abstract

We propose a new algorithm for solving the semiclassical time–dependent Schrödinger equation. The algorithm is based on semiclassical wavepackets. Convergence is proved to be quadratic in the time step and linear in the semiclassical parameter ε. To the the authors' best knowledge, all earlier algorithms had errors that grew as the semiclassical parameter was decreased.

1   Introduction

We consider the semiclassical time–dependent Schrödinger equation

$$i\,\varepsilon^2\,\partial_t \psi \;=\; H(\varepsilon)\,\psi\,, \tag{1}$$

where $\psi = \psi(x,\,t)$ is the wave function that depends on the spatial variables $x = (x_1,\ldots,x_d) \in \mathbb{R}^d$ and the time variable $t \in \mathbb{R}$. The Hamiltonian

$$H(\varepsilon) \;=\; -\,\frac{\varepsilon^4}{2}\,\Delta_x \;+\; V(x)$$

involves the Laplace operator Δ_x and a smooth real potential V.

The main challenges in the numerical solution of (1) result from the possibly high dimension d and the existence of several time– and spatial–scales governed by the small parameter ε. In chemical applications the ε represents the fourth root of the ratio between the electron mass and the mass of the lightest nucleus in the molecule. For instance, the $H + H_2$ reaction has $\varepsilon \approx 0.1528$; CO_2 has $\varepsilon \approx 0.0821$; while IBr is modeled by a one-dimensional system with $\varepsilon \approx 0.0511$. We note that ε has a clear fixed physical meaning, but if we regard the equation (1) as a whole range of models varying with the model-parameter ε, we recover the full quantum dynamics when $\varepsilon = 1$ and classical mechanics in the limit $\varepsilon \to 0$.

Here, we ignore the difficulties that arise from the possibly large dimension d and focus on the challenges caused by a small ε in the time-integration schemes. The preferred numerical integration scheme in quantum dynamics is the split operator technique [5] which, unlike Chebyshev or Lanczos schemes, does not suffer from a time step restriction, such as $\Delta t = O(\Delta x^2)$; see [8]. However, in the case of a semiclassical model (1), it is proved in [2] that the Lie-Trotter splitting requires a time-step of the order of ε^2, and the error is of order $\Delta t/\varepsilon^2$. For the Strang–splitting, convergence of order $(\Delta t)^2/\varepsilon^2$ was observed in [2, 1] and proved in [3]. Our own numerical experiments with a fourth-order splitting in time, together with spectral discretizations in space show the same factor of $1/\varepsilon^2$.

The small parameter ε forces the choice of a small time-step (and for a Fourier based space discretization, a huge number of grid points) in order to have reliable results, even for a fourth-order scheme. Recent research has been done to control the error in such time-splitting

spectral approximations [7]. We propose below a new splitting that is more appropriate for the semiclassical situation. We prove it has convergence of order $\varepsilon (\Delta t)^2$, which improves instead of deteriorating when ε is reduced. We are not aware of any other algorithm with a similar property.

The new integration scheme is based on a spatial discretization via semiclassical wavepackets [6]. The wavepackets have already been useful in the time integration of the semiclassical time-dependent Schrödinger equation in many dimensions via a special Strang–splitting [4].

The main idea of the time-integration scheme in [4] was Strang–splitting between the kinetic and potential energy, together with the observation that the kinetic part and a quadratic part of the potential can be integrated exactly. This yielded a second splitting of the potential into a quadratic part and a remainder. The line of attack in [6] was slightly different: an approximate solution was built upon the integration of a system of ordinary differential equations for the parameters of the wavepackets. Then a second system of ordinary differential equations was used for determining the coefficients of the wavepackets. Below, we present an algorithm that combines both of these two important ideas.

Let us start with a short introduction to semiclassical wavepackets. They are an example of a spectral basis consisting of functions that are defined on unbounded domains. For simplicity, we describe only the case of dimension $d = 1$, while the whole analysis can be carried out in general dimensions.

Given a set of parameters q, p, Q, P, a family of semiclassical wavepackets $\{\varphi_k^\varepsilon[q, p, Q, P], \ k = 0, 1, \ldots\}$ is an orthonormal basis of $L^2(\mathbb{R})$ that is constructed in [6] from the Gaussian

$$\varphi_0^\varepsilon[q, p, Q, P](x) \ = \ (\pi)^{-\frac{1}{4}} (\varepsilon Q)^{-\frac{1}{2}} \exp\left(\frac{i}{2\varepsilon^2} PQ^{-1}(x - q)^2 + \frac{i}{\varepsilon^2} p(x - q) \right),$$

via a raising operator. In the notation used here, Q and P correspond to A and iB of [6], respectively. Note that Q and P must obey the compatibility condition $\overline{Q}\, P - \overline{P}\, Q = 2\, i$; see [6]. Each state $\varphi_k(x) = \varphi_k^\varepsilon[q, p, Q, P](x)$ is concentrated in position near q and in momentum near p with uncertainties $\varepsilon|Q|\sqrt{k + \frac{1}{2}}$ and $\varepsilon|P|\sqrt{k + \frac{1}{2}}$, respectively. The recurrence relation

$$Q\sqrt{k + 1}\, \varphi_{k+1}^\varepsilon(x) \ = \ \frac{\sqrt{2}}{\varepsilon}\, (x - q)\, \varphi_k^\varepsilon(x) \ - \ \overline{Q}\sqrt{k}\, \varphi_{k-1}^\varepsilon(x)$$

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holds for all values of x. We gather together the parameters of the semiclassical wavepackets and write $\Pi(t) = (q(t), p(t), Q(t), P(t))$, so that $\varphi_k[\Pi](x) = \varphi_k^\varepsilon[q, p, Q, P](x)$.

We assume we have an initial condition $\psi(0)$ that is given as a linear combination of semiclassical wavepackets

$$\psi(0) = e^{iS(0)/\varepsilon^2} \sum_{k=0}^{K-1} c_k(0, \varepsilon) \varphi_k[\Pi(0)].$$

Note that we have an overall phase parameter $S(t)$ that will enlarge the parameter set. Hence, we shall also write $\varphi_k[\Pi, S] = e^{iS/\varepsilon^2}\varphi_k[\Pi]$. Note that K can be taken as large as we wish, just by inserting more trivial coefficients $c_k(0, \varepsilon) = 0$. Theorem 2.10 in [6] establishes an upper bound for the semiclassical approximation: If the potential $V \in C^{M+2}(\mathbb{R})$ satisfies $-C_1 < V(x) < C_2 e^{C_3 x^2}$, there is an approximate solution $v(t)$ of the semiclassical time–dependent Schrödinger equation (1) such that for any $T > 0$ we have:

$$\|\psi(t) - v(t)\| \le \mathcal{C}(T)\, \varepsilon^M, \qquad \text{for all } t \in [0, T].$$

From now on, $\mathcal{C}$ will denote a generic constant, not depending on ε or any involved time-step. We also only consider potentials V that satisfy the above conditions. The approximate solution in Theorem 2.10 in [6] is defined as

$$v(t) = e^{iS(t)/\varepsilon^2} \sum_{k=0}^{K-1} c_k(t, \varepsilon) \varphi_k[\Pi(t)], \tag{2}$$

with the parameters $\Pi(t)$ and $S(t)$ given by the solution to the following system of ordinary differential equations

$$\begin{aligned}
\dot{q}(t) &= p(t) \\
\dot{p}(t) &= -V'(q(t)) \\
\dot{S}(t) &= \frac{1}{2}p(t)^2 - V(q(t)) \\
\dot{Q}(t) &= P(t) \\
\dot{P}(t) &= -V''(q(t))\, Q(t).
\end{aligned} \tag{3}$$

The coefficients $c_k(t, \varepsilon)$ obey a linear system of ordinary differential equations. A similar result is valid in higher dimensions; see Theorem 3.6 of [6]. The dependence of $\mathcal{C}$ on the

end-time T is difficult to assess, and the system for the coefficients $c_k(t, \varepsilon)$ is difficult to solve, so an alternative numerical scheme is necessary.

This approximation result motivates us to look for an algorithm that does not deteriorate as $\varepsilon \to 0$. We note that our algorithm will separately handle the approximation for the parameters $\Pi(t)$ and $S(t)$ and the wave packet coefficients $c_k(t, \varepsilon)$. This observation is essential for the construction of the algorithm.

2 Time-splittings

A starting point for an efficient time-integration is the scheme proposed in [4]: As in Strang–splitting, we decompose the Hamiltonian

$$H \;=\; T + U + W$$

into its kinetic part $T \;=\; -\dfrac{\varepsilon^4}{2}\Delta_x$ and its potential part $V(x) \;=\; U(q(t), x) + W(q(t), x)$, where $U(q(t), x)$ is the quadratic Taylor expansion of $V(x)$ around $q(t)$ and $W(q(t), x)$ is the corresponding remainder:

$$V(x) \;=\; U(q, x) + W(q, x) \;=\; V(q) + V'(q)(x - q) + \frac{1}{2} V''(q)(x - q)^2 + W(q, x)\,.$$

We call this algorithm the L–splitting for a time-step of length Δt:

Algorithm 1 (L–Splitting)

1. *Propagate the solution for time $\frac{1}{2}\Delta t$, using only T.*
2. *Propagate the solution for time Δt, using only U.*
3. *Propagate the solution for time Δt, using only W.*
4. *Propagate the solution for time $\frac{1}{2}\Delta t$, using only T.*

As shown in [4], the steps 1, 2, and 4 reduce to simple updates of the numerically propagated parameters $\tilde{\Pi}$ and $\tilde{S}$ (starting, of course from the given $\Pi(0)$ and $S(0)$). The step 3 keeps the parameters $\tilde{\Pi}$ and $\tilde{S}$ fixed and evolves the set of coefficients via the system of ordinary differential equations

$$i\varepsilon^2 \dot{\tilde{c}} \;=\; F\left[\tilde{\Pi}\left(\frac{\Delta t}{2}\right)\right] \tilde{c}, \qquad \text{for } t \in [0, \Delta t]\,, \tag{4}$$

with a $K \times K$ matrix $F[\tilde{\Pi}(\frac{\Delta t}{2})]$ whose entries are

$$F_{j,k}\left[\tilde{\Pi}\left(\frac{\Delta t}{2}\right)\right] = \int \overline{\varphi_j[\tilde{\Pi}\left(\frac{\Delta t}{2}\right)](x)} \, W\!\left(\tilde{q}\left(\frac{\Delta t}{2}\right), x\right) \varphi_k[\tilde{\Pi}\left(\frac{\Delta t}{2}\right)](x) \, dx \, .$$

As mentioned in the Introduction, the global convergence of this algorithm (measured by the L^2-error) is observed to be $(\Delta t/\varepsilon)^2$, which can also be proved as in [3].

A splitting of $H = T + V$ that is of order 4 is the Y–splitting; see [9].

Denoting $\theta = 1/(2 - 2^{1/3})$, we have:

Algorithm 2 (Y–Splitting)
1. *Propagate the solution for time $\theta\frac{1}{2}\Delta t$, using only T.*
2. *Propagate the solution for time $\theta\Delta t$, using only V.*
3. *Propagate the solution for time $(1 - \theta)\frac{1}{2}\Delta t$, using only T.*
4. *Propagate the solution for time $(1 - 2\theta)\Delta t$, using only V.*
5. *Propagate the solution for time $(1 - \theta)\frac{1}{2}\Delta t$, using only T.*
6. *Propagate the solution for time $\theta\Delta t$, using only V.*
7. *Propagate the solution for time $\theta\frac{1}{2}\Delta t$, using only T.*

One can use this also for our decomposition $H = T + U + W$:

Algorithm 3 (YL–splitting $H = T + U + W$)
1. *Propagate the solution for time $\theta\frac{1}{2}\Delta t$, using only T.*
2. *Propagate the solution for time $\theta\Delta t$, using only U.*
3. *Propagate the solution for time $\theta\Delta t$, using only W.*
4. *Propagate the solution for time $(1 - \theta)\frac{1}{2}\Delta t$, using only T.*
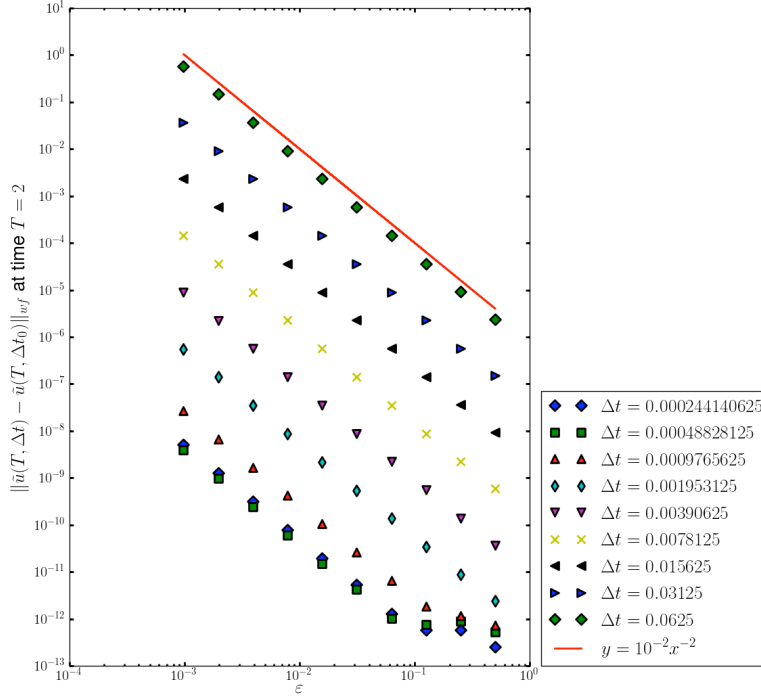5. *Propagate the solution for time $(1 - 2\theta)\Delta t$, using only U.*
6. *Propagate the solution for time $(1 - 2\theta)\Delta t$, using only W.*
7. *Propagate the solution for time $(1 - \theta)\frac{1}{2}\Delta t$, using only T.*
8. *Propagate the solution for time $\theta\Delta t$, using only U.*
9. *Propagate the solution for time $\theta\Delta t$, using only W.*
10. *Propagate the solution for time $\theta\frac{1}{2}\Delta t$, using only T.*

Figure 1: The error dependence on ε in the propagation with YL-splitting and wavepackets.

We observed convergence of order $(\Delta t^2/\varepsilon)^2$ in all tests based on this YL–splitting. Since the expensive propagation of the coefficients must be done three times (in the steps 3, 6, and 9) during each time step, the computational time is much greater than that for the L–splitting. We measure the error in the experiments via a numerical approximation of the L^2-norm based on 2^{16} equispaced points in the space domain; in order to emphasize that this is not the exact L^2-norm of the error, we denoted it by $\| \cdot \|_{wf}$; see Figure 1. The benchmark problem used in all numerical results presented here is based on the torsional potential $V(x) = 1 - \cos(x)$ with the initial value $\varphi_0[1, 0, 1, i]$ which is propagated from the initial time $t = 0$ to the end time $T = 2$. Tests with different potentials (including $V(x) = x^4$) and various other initial values produced similar results. In the results presented, we used $K = 8$ wavepackets. The equation (4) was solved via standard Padé approximation of the exponential matrix. We also used the Arnoldi method when computing with much larger K without observing any significant difference concerning the results in this paper. The components of $\tilde{F}\left[\left(\frac{\Delta t}{2}\right)\right]$ in (4) were computed via a very precise Gauss–Hermite quadrature

which was adapted to the shape of the wavepackets. In each case, the reference solution used to estimate the error was the numerical solution computed with the time step Δt_0 equal to half of the smallest time step displayed in the corresponding picture. Note that the solution based on traditional 2nd or 4th order splittings (Fourier or wavepacket based) cannot be used as a reference solution for the semiclassical splitting, since the errors grow like $1/\varepsilon^2$ as $\varepsilon \to 0$.

The critical idea for our new algorithm comes from the observation that for small ε, the largest errors arise from the computation of the parameters Π and S, while the expensive part of the computation is finding the coefficients c. Our algorithm combines the Strang–splitting for $H = (T + U) + W$ and the Y–splitting for $U + T$, $i.e.$, it is a 4th order scheme for the approximation of the parameters Π and S. We keep the expensive part of the computation (involving the remainder W) in the inner part of the Strang–splitting $H = \frac{1}{2}(T + U) + W + \frac{1}{2}(T + U)$ with a large time step Δt. We use a small time step δt for the cheap propagation of the parameters. This small time step is chosen so that the desired overall convergence rate $\varepsilon(\Delta t)^2$ is achieved with a minimum value of $\Delta t/\delta t = N = N(\varepsilon, \Delta t)$ small time steps per large time step. We call the resulting propagation algorithm semiclassical–splitting.

Algorithm 4 (Semiclassical–Splitting)

Define $N := \texttt{ceil}\left(1 + \dfrac{\sqrt{\Delta t}}{\varepsilon^{3/4}}\right)$ and $\delta t := \dfrac{\Delta t}{N}$, which hence satisfies

$\delta t \leq \min\left\{\varepsilon^{3/4}\sqrt{\Delta t},\ \Delta t\right\}$.

1. *Propagate the solution for time $\frac{1}{2}\Delta t$ via N steps of length δt using the Y–splitting for $T + U$ (Algorithm 2).*
2. *Propagate the solution for time Δt, using only W, i.e., solve equation (4).*
3. *Propagate the solution for time $\frac{1}{2}\Delta t$ via N steps of length δt using the Y–splitting for $T + U$ (Algorithm 2).*

Figures 2, 3, 4 display the results of numerical experiments based on this semiclassical splitting. They confirm our theoretical considerations from Section 3, as long as the strong round off in $e^{-i\alpha/\varepsilon^2} - e^{-i(\alpha+\mathrm{eps})/\varepsilon^2}$ (with $\alpha \neq 0$ and eps $=$ machine precision) in the measurement of the error can be avoided. The effect of this round off in the measurement of the error for Δt and ε simultaneously small is evident in Figure 2; this effect is missing when measuring the error in the coefficients; see Figure 3.

The adaptive choice of the inner time step δt is responsible for the complex behavior of the time-convergence curves in Figure 4: from order 4 for large ε and small Δt to only order 2 for large Δt. A similar picture arises if we study the convergence for observables.
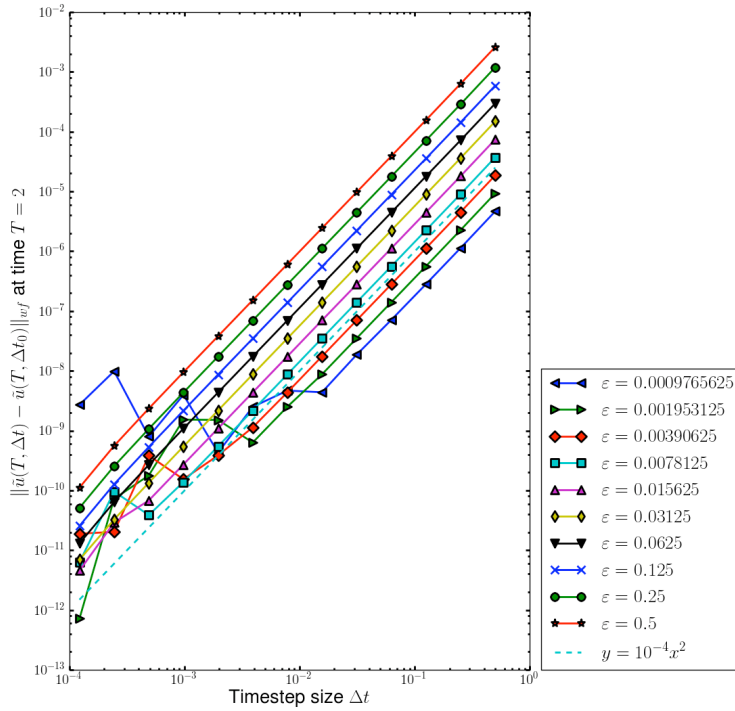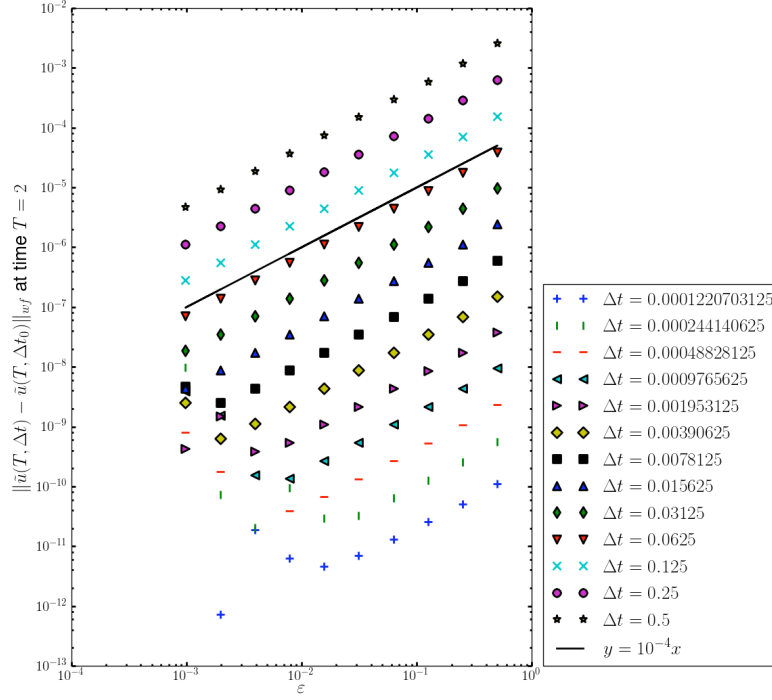
Figure 2: The error dependence on ε and on Δt (semiclassical–splitting).
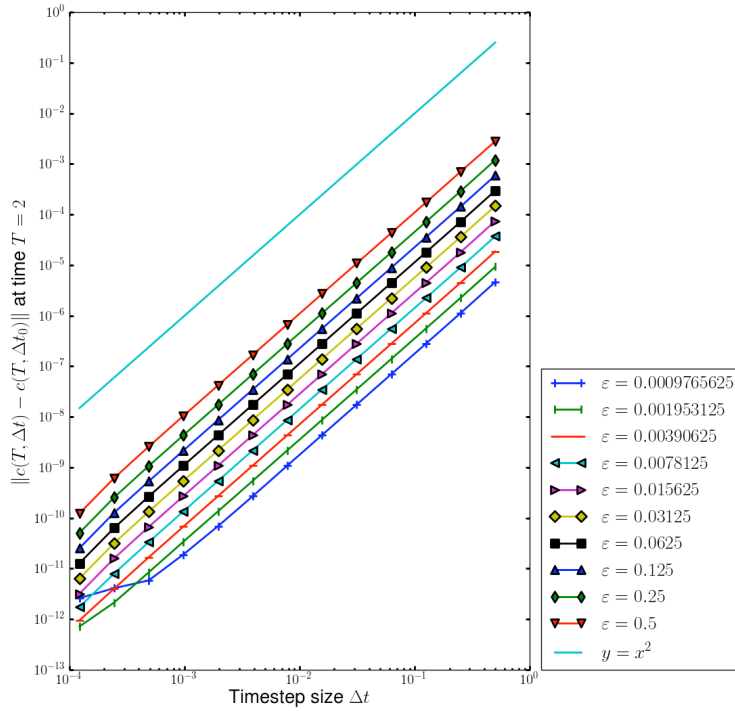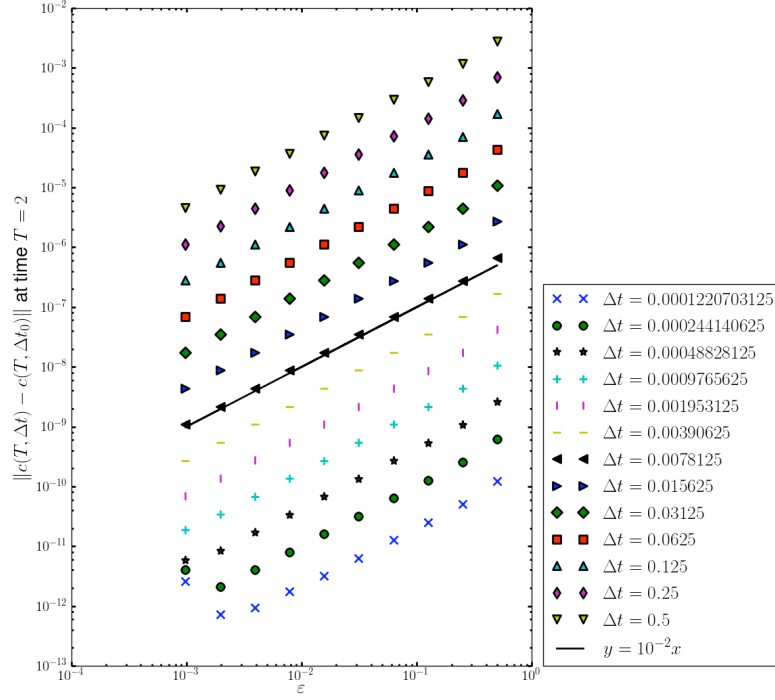
Figure 3: The error dependence on ε and on Δt of the coefficients (semiclassical–splitting).
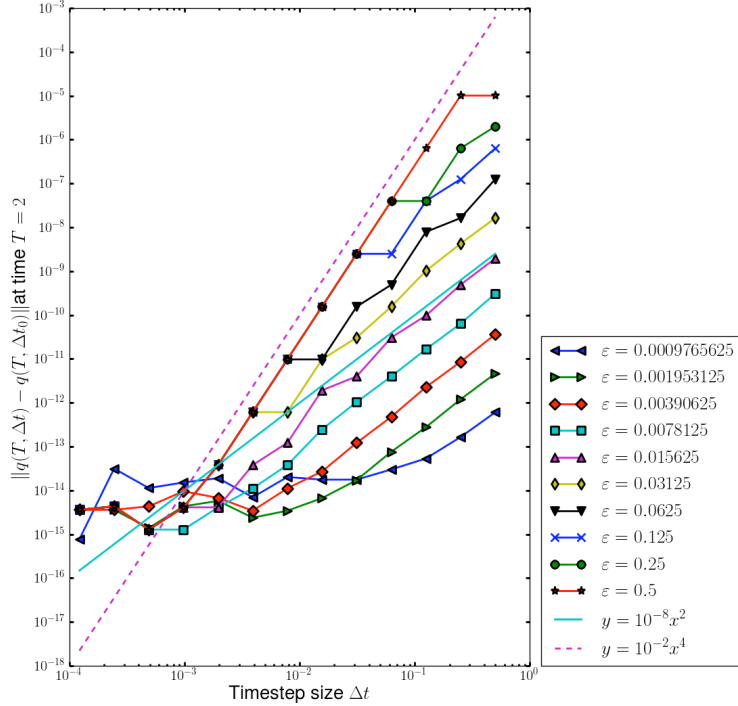
Figure 4: Time convergence of the parameter q (semiclassical–splitting).

3 Convergence Results for the Semiclassical–Splitting Algorithm

We first note that one cannot address the convergence of the proposed algorithms via the local error representation of exponential operator splitting methods as in Section 5 of [3]. This is because the parameters ε and Δt enter the splitting in fundamentally different ways.

The overall error when using the semiclassical–splitting algorithm consists of an approximation error caused by using the representation with a finite number of moving wavepackets and a time-discretization error. Our main result is the following:

Theorem 1. *Suppose the initial value is*

$$\psi(0) \;=\; e^{iS(0)/\varepsilon^2} \sum_{k=0}^{K_1-1} c_k(0, \, \varepsilon) \, \varphi_k[\Pi(0)] \,,$$

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and that $K \geq K_1 + 3(N - 1)$, where $N = 1$, 2, or 3. If the potential $V \in C^5(\mathbb{R})$ and its derivatives satisfy $-C_1 < V^{(s)}(x) < C_2 e^{C_3 x^2}$, for $s = 0$, 1, $\ldots$, 5, then there are constants $\mathcal{C}_1$ and $\mathcal{C}_2$, such that

$$\| \psi(t) - \tilde{u}(t) \| \ \leq \ \mathcal{C}_1 \, \varepsilon^N \ + \ \mathcal{C}_2 \, \varepsilon \, (\Delta t)^2 \,, \tag{5}$$

where $\tilde{u}$ is constructed via the semiclassical splitting Algorithm 4. The constants $\mathcal{C}_1$ and $\mathcal{C}_2$ do not depend on Δt, δt, or ε, but depend on the potential V and its derivatives up to 5th order, on the sup's of $|Q|$, $|\dot{Q}|$, $|\ddot{Q}|$, $|q|$, $|\dot{q}|$, $|\ddot{q}|$ from (3) on $[0, T]$, on the number K of wavepackets used in the approximation, and on the final integration time T.

Remark If $V \in C^l(\mathbb{R})$ with $l > 5$, then the theorem is true with a larger N. More precisely, if $K \geq K_1 + 3(N - 1)$, the first term on the right hand side of (5) still has the form $\mathcal{C}_1 \, \varepsilon^N$, but the restriction on N becomes $N = 1$, 2, $\ldots$, $l - 2$, and $\mathcal{C}_1$ depends also on $V^{(s)}$ for $s \leq l$.

We prove this theorem in several steps. First, we introduce an approximation to the solution of the time dependent Schrödinger equation u similar to (2). Here, the parameters Π and S satisfy the system (3), and the coefficients c_k are given by the solution to the linear system of ordinary differential equations

$$i \, \varepsilon^2 \, \dot{c} \ = \ F \left[\Pi(t) \right] \, c \,, \tag{6}$$

where the $K \times K$ matrix $F[\Pi]$ has entries

$$F_{j,k} \left[\Pi(t) \right] \ = \ \int \overline{\varphi_j[\Pi(t)](x)} \ W(q(t), x) \ \varphi_k[\Pi(t)](x) \, dx \,.$$

Theorem 2. *The approximation error is bounded by the Galerkin error:*

$$\| \psi(T) - u(T) \| \ \leq \ \frac{1}{\varepsilon^2} \int_0^T \ \| P_K W u - W u \| \, dt \,.$$

Under the hypotheses of Theorem 1 (or the remark after it), the integrand here is bounded by

$$\mathcal{C}_{K,Q,W} \, \varepsilon^{N+2} \,.$$

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Proof. Corollary 2.6 of [6] ensures that each $\varphi_k[\Pi, S]$ satisfies the time dependent Schrödinger equation with the Hamiltonian $T + U$. Using (6), we see that

$$i\,\varepsilon^2\,\partial_t u \;=\; H(\varepsilon^2)\,u \;+\; P_K\,W u \;-\; W u\,,$$

where P_k denotes the L^2-orthogonal projection into the space spanned by the basis functions $\varphi_0[\Pi], \ldots, \varphi_{K-1}[\Pi]$.

We intend to apply the elementary Lemma 2.8 of [6], so we only have to find an upper bound for the Galerkin error

$$\|P_K W u - W u\|\,.$$

We note that $\|P_K W u - W u\|$ is decreasing in K, so when $N = 1$, it suffices to prove $\|P_{K_1} W u - W u\| = O(\varepsilon^3)$. However, $\|P_K W u - W u\| \le \|W u\|$, and since W is locally cubic near $x = q$, estimate (2.68) of [6] immediately proves the result.

When $N = 2$, it suffices to prove $\|P_{K_1+3} W u - W u\| = O(\varepsilon^4)$. To show this, we first note that $\|(1 - P_{K_1})u\| = O(\varepsilon)$ as in the proof of Theorem 2.10 of [6]. Next,

$$W(x,\,t) \;=\; V'''(q(t))\,(x - q(t))^3 \;+\; V''''(\xi(x,\,t))\,(x - q(t))^4.$$

We have

$$
\begin{aligned}
\|(P_{K_1+3} - 1)\,W\,u\| \;&\le\; \|(P_{K_1+3} - 1)\,W\,P_{K_1}\,u\| \\
&\quad +\; \|(P_{K_1+3} - 1)\,W\,(1 - P_{K_1})\,u\| \\
&\le\; \|(P_{K_1+3} - 1)\,V''''(q)\,(x - a)^3\,P_{K_1}\,u\| \\
&\quad +\; \|(P_{K_1+3} - 1)\,V''''(\xi(x,\,t))\,(x - q)^4\,P_{K_1}\,u\| \\
&\quad +\; \|(P_{K_1+3} - 1)\,W\,(1 - P_{K_1})\,u\|.
\end{aligned}
$$

The first term on the right hand side is zero since $(P_{K_1+3} - 1)\,(x - q)^3\,P_{K_1} = 0$. The second term is $O(\varepsilon^4)$ as in the proof of Theorem 2.10 of [6]. The third term is bounded by $\|W\,(1 - P_{K_1})\,u\|$. This quantity is $O(\varepsilon^4)$ because W is locally cubic as in the $N = 1$ case and $\|(1 - P_{K_1})\,u\| = O(\varepsilon)$.

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The $N = 3$ case is similar, but with the next order Taylor series estimates. The $N \leq l - 2$ case is similar under the hypotheses of the remark, but with higher order Taylor series estimates. $\square$

The first term on the right hand side of the estimate in *Theorem 1* arises from the estimate in *Theorem 2*. The rest of this paper concerns the second term.

We concentrate on the local time-error when approximating $u(\Delta t)$; if we show that it can be bounded by $\varepsilon \, (\Delta t)^3$, then standard arguments and *Theorem 2* prove *Theorem 1*.

The first step in *Algorithm 4* produces approximations $\tilde{\Pi}\left(\frac{\Delta t}{2}\right)$ of $\Pi\left(\frac{\Delta t}{2}\right)$ and $\tilde{S}\left(\frac{\Delta t}{2}\right)$ of $S\left(\frac{\Delta t}{2}\right)$, both with errors $\mathcal{C} \, (\delta t)^4 \, \frac{\Delta t}{2}$. Given these approximate parameters, the coefficients $\tilde{c}_k$ solve the system (6) with the constant matrix $\tilde{F} = F[\tilde{\Pi}\left(\frac{\Delta t}{2}\right)]$ on right hand side, as in (4). They enter in the expression of the numerical solution at the end of the Strang–splitting

$$\tilde{u}(\Delta t) \;=\; e^{i \tilde{S}(\Delta t)/\varepsilon^2} \sum_{k=0}^{K-1} \tilde{c}_k(\Delta t, \, \varepsilon) \; \varphi_k[\tilde{\Pi}(\Delta t)] \,.$$

In practice, they are computed via expensive (Padé or Arnoldi) iterations, but here we assume they are computed exactly.

In order to shorten the formulas, we denote $\varphi_k = \varphi_k[\Pi]$ and $\tilde{\varphi}_k = \varphi_k[\tilde{\Pi}]$.

To facilitate the proof, we introduce $u_1(t)$, constructed via the parameters $\Pi(t)$ from the exact solution of (3) and coefficients c^1 from the linear system with constant matrix $F^1 = F[\Pi^1]$, where $\Pi^1 = \Pi\left(\frac{\Delta t}{2}\right)$:

$$i \, \varepsilon^2 \, \dot{c}^1 \;=\; F^1 \, c^1 \,, \qquad \text{for } t \in [0, \, \Delta t] \,.$$

With this we construct

$$u_1(t) \;=\; e^{i S(t)/\varepsilon^2} \sum_{k=0}^{K-1} c_k^1(t, \, \varepsilon) \; \varphi_k[\Pi(t)] \,,$$

which has the parameters of u, but the coefficients c^1. The local error then decomposes as

$$\|u(\Delta t) - \tilde{u}(\Delta t)\| \;\leq\; \|u(\Delta t) - u_1(\Delta t)\| \;+\; \|u_1(\Delta t) - \tilde{u}(\Delta t)\| \,.$$

The next two theorems give upper bounds for these two last terms, ensuring the desired bound on the local error.

We now take a more careful look at the Galerkin matrix F. A crucial observation is that as in section 4.1 of [4], the change of variables $x = q + \varepsilon y$ allows us to represent

$$F_{j,k} = \int \overline{\phi_j(y)}\, W(q + \varepsilon y)\, \phi_k(y)\, dy \tag{7}$$

in terms of ε–independent, but orthogonal functions ϕ_k, given by the recurrence relation

$$\phi_0(y) = \pi^{-1/4}\, |Q|^{-1/2}\, e^{-|Q|^{-2}|y|^2/2},$$

$$Q\sqrt{k_j+1}\, \phi_{k+1}(y) = \sqrt{2}\, y\, \phi_k(y) - \overline{Q}\,\sqrt{k_j}\, \phi_{k-1}(y)\,.$$

By this change of variables, all of the dependence on ε has been moved out of the wavepackets and put into the operator $W(q + \varepsilon y)$, which is

$$W(q + \varepsilon y)$$
$$= V(q(t) + \varepsilon y) - V(q(t)) - V'(q(t))\,\varepsilon\, y - \frac{1}{2} V''(q(t))\,\varepsilon^2\, y^2 \tag{8}$$
$$= \frac{1}{6} V'''(\zeta(y))\,\varepsilon^3\, y^3$$
$$= \int_{q(t)}^{q(t)+\varepsilon y} \frac{(q(t) + \varepsilon y - z)^2}{2}\, V'''(z)\, dz\,. \tag{9}$$

Note that Q is not present in the expression for W, while the functions ϕ_k are wavepackets that depend on Q only: $\phi_k = \varphi_k^{\varepsilon=1}\left[0, 0, Q, i\,(\overline{Q})^{-1}\right]$. We use this observation in the proofs of the following lemmas.

Lemma 1. *Suppose g and Z are functions on $\mathbb{R}$ that satisfy $g \in L^2(\mathbb{R},\, dy)$ and $|Z(y)| \leq P(y)\, e^{C\varepsilon^2 y^2/2}$, where P is a non-negative polynomial.*
Then there exists a constant $\mathcal{C}$ depending only on j and Q, such that

$$|\langle \phi_j,\, Zg \rangle| = \left| \int \overline{\phi_j(y)}\, Z(y)\, g(y)\, dy \right| \leq \mathcal{C}\, \|g\|\,, \tag{10}$$

for all $\varepsilon < \dfrac{|Q|^{-1}}{\sqrt{2\,C}}$.

Proof. By the Schwarz inequality, $|\langle \phi_j,\, Z\,g \rangle| \leq \|Z\,\phi_j\|\,\|g\|$, so it suffices to prove that $\|Z\,\phi_j\|$ is finite. However,

$$\|Z\,\phi_j\|^2 = \pi^{-1/2}\, |Q|^{-1} \int_{\mathbb{R}} |Z(y)|^2\, |p(y)|^2\, e^{-y^2/|Q|^2}\, dy\,,$$

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where p is a polynomial. Under our assumptions, elementary estimates show that this integral is bounded by a constant, uniformly for $\varepsilon^2 < \frac{|Q|^{-2}}{2C}$. $\qquad\square$

Lemma 2. *The entries of the matrix $F[\Pi]$ and their first two time-derivatives are bounded by constants times ε^3:*

$$|F_{j,k}(t)| \;\leq\! \mathcal{C}_0\, \varepsilon^3, \quad |\dot{F}_{j,k}(t)| \;\leq\! \mathcal{C}_1\, \varepsilon^3, \quad and \quad |\ddot{F}_{j,k}(t)| \;\leq\! \mathcal{C}_2\, \varepsilon^3,$$

where the constants $\mathcal{C}_l$ depend only on j, k, Q, $\dot{Q}$, $\ddot{Q}$, $\dot{q}$, $\ddot{q}$, and bounds on the third, fourth, and fifth derivatives of V.

Proof. We use expression (9) to write

$$F_{j,k}(t) \;=\; \int_{\mathbb{R}} \overline{\phi_j(y)} \int_{q(t)}^{q(t)+\varepsilon y} \frac{(q(t) + \varepsilon y - z)^2}{2}\, V'''(z)\; dz\; \phi_k(y)\; dy\,.$$

We then let $z = q(t) + \sigma \varepsilon y$ and rewrite the inner integral as

$$\int_0^1 \frac{(\varepsilon y\,(1-\sigma))^2}{2}\, V'''(q(t) + \sigma \varepsilon y)\; \varepsilon y\; d\sigma\,.$$

Next, we interchange the order of integration to obtain

$$F_{j,k}(t) \;=\; \varepsilon^3 \int_0^1 \frac{(1-\sigma)^2}{2} \int_{\mathbb{R}} \overline{\phi_j(y)}\; y^3\; V'''(q(t) + \sigma \varepsilon y)\; \phi_k(y)\; dy\; d\sigma\,. \qquad (11)$$

Lemma 1 with $Z(y) = y^3\, V'''(q(t) + \sigma \varepsilon y)$ gives the result for $F_{j,k}$.

To study $\dot{F}_{j,k}$, we take the time derivative of expression (11). The only time dependent quantities here are ϕ_j, ϕ_k, and $V'''(q(t) + \sigma \varepsilon y)$. The time dependence in the ϕ's comes only from $Q(t)$ and its conjugate, while the time dependence in $V'''(q(t) + \sigma \varepsilon y)$ comes only from $q(t)$. The result for $\dot{F}_{j,k}$ then follows by several applications of *Lemma 1*.

The result for $\ddot{F}_{j,k}$ follows from the same arguments applied to the second time derivative of (11). $\qquad\square$

Corollary 1.

$$\| F(t) \| \;\leq\; \mathcal{C}\, \varepsilon^3, \quad \| \dot{F}(t) \| \;\leq\; \mathcal{C}\, \varepsilon^3, \quad and \quad \| \ddot{F}(t) \| \;\leq\; \mathcal{C}\, \varepsilon^3,$$

where $\mathcal{C}$ has the same dependency as in Lemma 2 and depends additionally on K, but is independent of ε.

Lemma 3. *The error caused by using the approximate parameters $\tilde{\Pi}$ in the wavepacket φ_k satisfies*

$$\| \varphi_k(\Delta t) - \tilde{\varphi}_k(\Delta t) \| \;\leq\; \mathcal{C}\, \frac{(\delta t)^4}{\varepsilon^2}\, \Delta t\,.$$

Proof. Using a homotopy between Π and $\tilde{\Pi}$ one can prove[1] via careful calculations that

$$\| \varphi - \tilde{\varphi} \| \;\leq\; \mathcal{C}\, \frac{1}{\varepsilon^2}\, \Big(|q - \tilde{q}| + |p - \tilde{p}| + |Q - \tilde{Q}| + |P - \tilde{P}| \Big)\,.$$

The fact that the Y–splitting with time step δt for Π on an interval of length Δt is of fourth order then concludes this proof. $\square$

Lemma 4. *The error caused by using the approximate parameters $\tilde{\Pi}$ in the matrix F satisfies*

$$\left\| F^1 - \tilde{F} \right\| \;\leq\; \mathcal{C}\, (\delta t)^4\, \Delta t\,,$$

with the constant $\mathcal{C}$ depending on Q^1, $\tilde{Q}$, and V and its derivatives up to 3rd order.

Proof. The representation (7) of the entries of the matrix F that depends only on Q is again the key idea: It allows us to write $F^1_{j,k} - \tilde{F}_{j,k}$ as the sum of the following three terms

$$\left\langle \phi^1_j,\, W^1(\phi^1_k - \tilde{\phi}_k) \right\rangle \;+\; \left\langle \phi^1_j,\, (W^1 - \tilde{W})\, \tilde{\phi}_k) \right\rangle \;+\; \left\langle \phi^1_j - \tilde{\phi}_j,\, \tilde{W}\, \tilde{\phi}_k \right\rangle\,,$$

where

$$W^1(y) \;=\; W(q^1 + \varepsilon y) \;=\; \frac{1}{6}\, V'''(q^1 + \varepsilon \zeta^1(y))\, \varepsilon^3\, y^3$$

and

$$\tilde{W}(y) \;=\; W(\tilde{q} + \varepsilon y) \;=\; \frac{1}{6}\, V'''(\tilde{q} + \varepsilon \tilde{\zeta}(y))\, \varepsilon^3\, y^3\,.$$

Note that $\phi^1_j = \phi_j[Q]$ and $\tilde{\phi}_j = \phi_j[\tilde{Q}]$ do not depend on q, p, or ε. *Lemma 1* now applies with $Z = W^1/\varepsilon^3$ and $g = \phi^1_k - \tilde{\phi}_k$ or $Z = \tilde{W}/\varepsilon^3$ and $g = \phi^1_j - \tilde{\phi}_j$ to estimate the first and the last term, respectively, by $\mathcal{C}\varepsilon^3 \|\phi^1_k - \tilde{\phi}_k\|$ or $\mathcal{C}\varepsilon^3 \|\phi^1_j - \tilde{\phi}_j\|$. As in the previous lemma, the last terms are both bounded by $\mathcal{C}\,\varepsilon\,(\delta t)^4\,\Delta t$ which is one order (in ε) smaller that the stated result. The largest error arises from the middle term. We bound it using (8) with the corresponding q^1 and $\tilde{q}$ and *Lemma 1* again. This yields a bound of $\mathcal{C}\,(2 + \varepsilon + \frac{1}{2}\varepsilon^2)\,|q^1 - \tilde{q}|$. Combining all these estimates, we get the upper bound for the quantity in the lemma:

$$\left\| F^1 - \tilde{F} \right\| \;\leq\; \mathcal{C}\, \left((2 + \varepsilon + \frac{1}{2}\varepsilon^2)\, |q - \tilde{q}| + \varepsilon\, |Q - \tilde{Q}| \right),$$

which is bounded by $\mathcal{C}\,(\delta t)^4\,\Delta t$ as in the previous lemma. $\square$

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[1] Private communication from G. Kallert (now Gauckler) in 2010.

Theorem 3. *The difference between the approximate solution and the intermediate solution is*

$$\|u(\Delta t) - u_1(\Delta t)\| \leq \mathcal{C} \, \varepsilon \, (\Delta t)^3 \, ,$$

where $\mathcal{C}$ depends on K and V and its derivatives up to 5th order, on Q for t between 0 and T, but is independent of ε and Δt.

Proof. Denoting the (unitary) propagator for (6) by $\mathcal{U}(t, s)$, we can express the difference between the approximate solution and the intermediate solution as

$$\left\| \mathcal{U}(\Delta t, 0) \, c(0) - e^{-i \frac{\Delta t}{\varepsilon^2} F^1} \, c(0) \right\| = \left\| c(0) - \mathcal{U}(0, \Delta t) \, e^{-i \frac{\Delta t}{\varepsilon^2} F^1} \, c(0) \right\| \, .$$

We abbreviate $F(s) := F[\Pi(s)]$, and observe that since $F^1 = F[\Pi\left(\frac{\Delta t}{2}\right)]$, the expression in the second norm above is the integral from 0 to Δt of

$$\mathcal{U}(0, s) \frac{1}{\varepsilon^2} \left(F(s) - F^1 \right) e^{-i s F^1/\varepsilon^2} \, c(0).$$

Thus, we have

$$\| u(\Delta t) - u_1(\Delta t) \| = \frac{1}{\varepsilon^2} \left\| \int_0^{\Delta t} \mathcal{U}(0, s) \left(F(s) - F^1 \right) e^{-i s F^1/\varepsilon^2} \, c(0) \, ds \right\| \, .$$

Standard arguments from the proof of the convergence order of the midpoint quadrature rule then shows that

$$\| u(\Delta t) - u_1(\Delta t) \| \leq \frac{(\Delta t)^3}{\varepsilon^2} \, \|R\|$$

with the remainder R involving the Peano kernel and a factor containing the second derivative with respect to s of the integrand in the above formula. The first derivative of the integrand is

$$\mathcal{U}(0, s) \frac{i}{\varepsilon^2} F(s) \left(F(s) - F^1 \right) e^{-i s F^1/\varepsilon^2} \, c(0)$$
$$+ \, \mathcal{U}(0, s) \, \dot{F}(s) \, e^{-i s F^1/\varepsilon^2} \, c(0)$$
$$+ \, \mathcal{U}(0, s) \left(F(s) - F^1 \right) \frac{-i}{\varepsilon^2} F^1 \, e^{-i s F^1/\varepsilon^2} \, c(0)$$

The second derivative has an even longer expression, but has the same character as the first one: every term containing the factor $1/\varepsilon^2$ contains also a factor of F^1 or $F(s)$ or $\dot{F}(s)$,

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which are of order ε^3, according to *Corollary 1*. Hence, the leading order terms are those involving only $\dot{F}(s)$ and $\ddot{F}(s)$, *i.e.*, similar to the middle term in the first derivative. Those terms are themselves of order ε^3, which shows that the remainder R is bounded by $\mathcal{C}\,\varepsilon^3$. $\quad\square$

Theorem 4. *The difference between the intermediate solution and the numerical solution is*

$$\| u_1(\Delta t) - \tilde{u}(\Delta t) \| \;\leq\; \mathcal{C}\,\frac{(\delta t)^4}{\varepsilon^2}\,\Delta t\,.$$

Proof. By the triangle inequality,

$$
\begin{aligned}
&\| u_1(t) - \tilde{u}(t) \| \\
&\leq\; \left| e^{-\,i\,S(t)/\varepsilon^2} - e^{-\,i\,\tilde{S}(t)/\varepsilon^2} \right| \;+\; \left\| \sum_{k=0}^{K-1} c_k^1(t)\,\varphi_k(t) \;-\; \tilde{c}_k(t)\,\tilde{\varphi}_k(t) \right\| \\
&\leq\; \mathcal{C}\,\frac{|S(t) - \tilde{S}(t)|}{\varepsilon^2} \;+\; \left\| \sum_{k=0}^{K-1} c_k^1(t)\,(\varphi_k(t) - \tilde{\varphi}_k(t)) \right\| \;+\; \| \mathbf{c}^1(t) - \tilde{\mathbf{c}}(t) \| \\
&\leq\; \mathcal{C}\,\frac{|S(t) - \tilde{S}(t)|}{\varepsilon^2} \;+\; \sqrt{\sum_{k=0}^{K-1} \|\varphi_k - \tilde{\varphi}_k\|^2} \;+\; \| \mathbf{c}^1(t) - \tilde{\mathbf{c}}(t) \|\,.
\end{aligned}
$$

We rewrite equation (4) as

$$i\,\varepsilon^2\,\dot{\tilde{c}} \;=\; F_1\,\tilde{c} \;+\; (\tilde{F} - F_1)\,\tilde{c}\,, \qquad \text{for} \quad t \in [0, \Delta t]\,.$$

Lemma 4 and elementary Lemma 2.8 of [6] give $\|\mathbf{c}(t) - \tilde{\mathbf{c}}(t)\| \leq \mathcal{C}\,\frac{(\delta t)^4}{\varepsilon^2}\,(\Delta t)^2$. *Lemma 3* and the error in the splitting of (3) then yield the result. $\quad\square$

Finally, *Theorems 3, 4*, and the triangle inequality give us an estimate of the local error:

$$\| u(\Delta t) - \tilde{u}(\Delta t) \| \;\leq\; \mathcal{C}\,\frac{(\delta t)^4}{\varepsilon^2}\,(\Delta t) \;+\; \mathcal{C}\,\varepsilon\,(\Delta t)^3 \;\leq\; \mathcal{C}\,\varepsilon\,(\Delta t)^3$$

if we choose

$$\delta t \;\leq\; \varepsilon^{3/4}\,\sqrt{\Delta t}\,.$$

This shows that the number N of time-steps for solving (3) via a splitting is at least $N \geq \sqrt{\Delta t}\,\varepsilon^{-3/4}$ for the semiclassical splitting *Algorithm 4*. With such a choice of N, standard arguments and *Theorem 2* imply the result on the global error in our main *Theorem 1*.

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