Analysis of the Toolkit method for the time-dependent Schrödinger equation

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ANALYSIS OF THE "TOOLKIT" METHOD FOR THE TIME-DEPENDENT SCHRÖDINGER EQUATION

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Abstract. The goal of this paper is to provide an analysis of the "toolkit" method used in the numerical approximation of the time-dependent Schrödinger equation. The "toolkit" method is based on precomputation of elementary propagators and was seen to be very efficient in the optimal control framework. Our analysis shows that this method provides better results than the second order Strang operator splitting. In addition, we present two improvements of the method in the limit of low and large intensity control fields.

Keywords : Toolkit method, quantum control, time-dependent Schrödinger equation.
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1. Introduction

The control of the evolution of molecular systems at the quantum level has been a long standing goal ever since the beginning of the laser technology. After an initial slowed down of the investigations in this area due to unsuccessful experiments, the realization that the problem can be recast and attacked with the tools of (optimal) control theory [1] greatly contributed to the first positive experimental results [2, 3, 4, 5, 6]. On the otherhand, exact [7] and approximate [8, 9] controllability results were obtained and provided a relevant mathematical framework. Ever since, the desire to understand theoretically how the laser acts to control the molecule lead the investigators to resort to numerical simulations which require repeated resolution of the Time Dependent Schrödinger Equation of the type (1); additional motivation comes from related contexts (online identification algorithms, learning algorithms, quantum computing [10], etc.).

The numerical method used to solve the time dependent Schrödinger equation must provide accurate results without prohibitive computational cost. The conservation of the L^2 norm of the wave function $\psi(x,t)$ is also generally required for stability and as a mean of qualitative validation of the numerical solution.

In this context, the second order Strang operator splitting is often considered [11, 12, 13]. However, this method suffers from two drawbacks. First, the numerical error is proportional to the norm of the control which implies poor accuracy when dealing with large laser fields $\varepsilon(t)$ and make necessary the use of small time steps. Secondly, it requires at each time step three matrix products. This difficulty is enhanced in some particular settings e.g., in optimal control, where the matrices involved in the control term must be assembled online.

Recently introduced, the "toolkit method" [14, 15] solves this last problem by precomputing a set of elementary matrices, used in the numerical resolution. Each matrix is associated to (one or several) field values and enables to solve the evolution over one time step. This algorithm has been used in various frameworks and shows excellent results. It has also been coupled successfully with optimal control and identification issues [16]. The dependence on the L^∞-norm of the control, which is a restriction of the Strang method, is also improved by the "toolkit method" as it will be shown in our analysis.

The goal of the paper is to provide a (first) numerical analysis of the "toolkit method". Our mathematical tools are related to that in [11] (but for a different setting; see also [17, 18] for connected results); the treatment here is different because of the quantization appearing in the values of the control $\varepsilon(t)$ which impacts both the mathematical analysis and the numerical efficiency of the method. The analysis enables us to propose two possible improvements.

The paper is organized as follows: after having introduced the model and some notations in Section 2, the "toolkit" method is presented and analyzed in Section 3. An improvement of this method in the limit of small control fields is introduced in Section 4. A second improvement, in the limit of large control fields is given in Section 5. Finally, Section 6 gathers some numerical results.

2. Model and notations

In this section, we present the Schrödinger Equation that will be considered in the paper and some useful notations. Note that the algorithms we consider in this paper actually apply for other types of Schrödinger equations. Indeed, they are rather based on the algebraic structure of the control problem (bilinear control) than on the regularity of the solution. In this way, the error estimates that we will obtain hold not only in L^2, but also in H^2.

We consider the time dependent Schrödinger equation (TDSE) ($\gamma \in \mathbb{N}$):

$$\left\{ \begin{array}{ll} i\partial_t \psi(x,t) = (H_0 - \mu(x)\varepsilon(t))\psi(x,t), & x \in \mathbb{R}^\gamma \\ \psi(x,0) = \psi_0(x), & x \in \mathbb{R}^\gamma. \end{array} \right. \tag{1}$$

This equation governs the evolution of a quantum system, described by its wave function ψ, that interacts with a laser pulse of amplitude ε, the control variable. The factor μ is the dipole moment operator of the system. The Hamiltonian of the system is $H_0 = -\Delta_x + V$ where Δ_x is the Laplacian operator over the space variables and $V = V(x)$ the electrostatic potential in which the system evolves. We refer to [19] for more details about models involved in quantum control. Note that to obtain Eq. (1), one has considered the laser effect as a perturbative term, so that the control term $\varepsilon(t)\mu(x)$ is obtained through a first order approximation with respect to $\varepsilon(t)$. While often considered, this approximation fails at describing some models involving non linear laser-dipole interaction, see e.g. [20]. Consequently, the norm of the field cannot be always considered as a small parameter, and numerical solvers have to tolerate large controls, as the one described here after.

Another distinct circumstance is when there are M systems which are exposed to the same laser field. Each system is characterized by its own internal Hamiltonian H_0^k and dipole moment $\mu_k(x)$; in addition each has its own orientation denoted ξ_k with respect to the incident laser direction. Some systems may be identical, in which case they will share the same H_0^k and $\mu_k(x)$ but may have different ξ_k. The governing equation is:

$$\left\{ \begin{array}{ll} i\partial_t \psi_k(x,t) = (H_0^k(x) - \mu_k(x)\cos(\xi_k)\varepsilon(t))\psi_k(x,t), & x \in \mathbb{R}^\gamma \\ \psi_k(x,0) = \psi_{k0}(x), & x \in \mathbb{R}^\gamma, k = 1, ..., M. \end{array} \right. \tag{2}$$

In this case the goal is to control all systems at the same time. We refer the reader to [21, 22, 23] for (positive) controllability results and numerical simulations performed with up to $M = 300$ systems or even in the continuous limit $\xi \in [-1, 1]$.

Throughout this paper, $T > 0$ is the time of control of a quantum system. The space $L^p(0,T;X)$, with $p \in [1,+\infty)$ denotes the usual Lebesgue space taking its values in a Banach space X. The notation $W^{1,1}(0,T)$ corresponds to the space of time dependent functions belonging to $L^1(0,T;\mathbb{R})$ such that their first time derivative also belongs to $L^1(0,T)$. We denote by L^2 the space $L^2(\mathbb{R}^\gamma,\mathbb{C})$ and by $W^{2,\infty}$ and H^2 the Sobolev spaces $W^{2,\infty}(\mathbb{R}^\gamma,\mathbb{R})$ and $H^2(\mathbb{R}^\gamma,\mathbb{C})$. The space $\mathcal{L}(H^2)$ is the space of linear functionals on H^2. One can refer to [24] (or the introduction of [25]) for more details about the definitions of these functional spaces.

Finally, in order to introduce some numerical solver of (1), let us consider an integer N and $\Delta t > 0$ such that $N\Delta t = T$. We introduce the time discretization $(t_j)_{0\leq j\leq N}$ of $[0,T]$ with $t_j = j\Delta t$ and we also denote by $t_{j+\frac{1}{2}}$ the intermediate time $\frac{t_j+t_{j+1}}{2} = (j+\frac{1}{2})\Delta t$.

Let us first recall some basic results of existence and regularity of the solution of the TDSE. These are corollaries of a general result on time dependent Hamiltonians (see [26], p. 285, Theorem X.70).

Lemma 1. *Let $\mu \in \mathcal{L}(H^2)$, $V \in W^{2,\infty}$, $\varepsilon \in W^{1,1}(0,T)$ and $\psi_0 \in H^2$. The Schrödinger equation*

$$\left\{ \begin{array}{ll} i\partial_t\psi(t) = (H_0 - \mu\varepsilon(t))\,\psi(t) & \mathbb{R}^\gamma \times (0,T) \\ \psi(0) = \psi_0, & \mathbb{R}^\gamma, \end{array} \right. \tag{3}$$

has a unique solution $\psi \in L^\infty(0,T;H^2) \cap W^{1,\infty}(0,T;L^2)$ such that

$$\|\psi(t)\|_{L^\infty(0,T;H^2)} + \|\partial_t\psi(t)\|_{L^\infty(0,T;L^2)} \leq C\left(1 + \|\mu\|_{\mathcal{L}(H^2)}\|\varepsilon\|_{W^{1,1}(0,T)}\right)\|\psi_0\|_{H^2}.$$

Moreover, for all $t \in [0,T]$, $\quad \|\psi(t)\|_{L^2} = \|\psi_0\|_{L^2}$.

It is also well known (see [25] for instance) that for any $T > 0$ and $\phi_0 \in H^2$, if we have $\varepsilon(t) = \bar{\varepsilon} \in \mathbb{R}$, independent of time t, the Schrödinger equation

$$\left\{ \begin{array}{ll} i\partial_t\phi(t) = (H_0 - \mu\bar{\varepsilon})\,\phi(t), & \mathbb{R}^\gamma \times (0,T) \\ \phi(0) = \phi_0, & \mathbb{R}^\gamma \end{array} \right.$$

has a unique solution $\phi(t) = S(t)\phi_0$ such that $\phi \in C([0,T];H^2) \cap C^1([0,T];L^2)$, where $(S(t))_{t\in\mathbb{R}}$ denotes the one-parameter semi-group generated by the operator $H_0 - \mu\bar{\varepsilon}$. Moreover, for all $t \in [0,T]$, $S(t) \in \mathcal{L}(H^2)$ and we have

$$\begin{array}{ll} S(t)\phi_0 \in C(0,T;H^2), & \forall\phi_0 \in H^2; \\ \|S(t)\|_{\mathcal{L}(H^2)} \leq 1 + CT \leq K, & \forall t \in [0,T]; K = K(\|\mu\|_{\mathcal{L}(H^2)}, \varepsilon_{\max}); \\ \|S(t)\phi_0\|_{L^2} = \|\phi_0\|_{L^2}, & \forall\phi_0 \in L^2, \forall t \in \mathbb{R}; \\ S(0) = \mathrm{Id}, & \\ S(t+s) = S(t)S(s), & \forall s,t \in \mathbb{R}. \end{array} \tag{4}$$

Therefore, the solution of Eq. (3) is obtained equivalently as a solution to the integral equation

$$\psi(t) = S(t)\psi_0 + i\int_0^t S(t-s)(\varepsilon(s) - \bar{\varepsilon})\mu\psi(s)\,ds.$$

3. The "toolkit" method

We now present the "toolkit" method and describe the corresponding error analysis.

3.1. **Algorithm.** In this method, we assume that the control field ε satisfies the following hypothesis:

$$(\mathcal{H}) \qquad \forall t \in [0,T],\ \varepsilon(t) \in [\varepsilon_{\min}, \varepsilon_{\max}].$$

The values of the control field are discretized according to:

$$\bar{\varepsilon}_\ell = \varepsilon_{\min} + \ell\Delta\varepsilon,\ \ell = 0...m, \tag{5}$$

with $m = \frac{\varepsilon_{\max}-\varepsilon_{\min}}{\Delta\varepsilon}$. Here, the values $\bar{\varepsilon}_\ell$ have been here uniformly chosen in the interval $[\varepsilon_{\min}, \varepsilon_{\max}]$. If some properties of the field are known, e.g. its mean value or its variance, some improvement of the method can be obtained by optimizing the distribution of the values $\bar{\varepsilon}_\ell$. More generally, this topic enters the field of scalar quantization, that will not be considered in this paper. We refer to [27] and the references therein for a review of standard methods in this domain.

Remark 1. *The hypothesis $\mathcal{H}$ often holds in practical cases. From the experimental point of view, there exists a technological bound for the laser amplitude. From the mathematical point of view, the field solves an optimality system of equations, which induces L^∞ bounds on the control. For example, consider the minimization of the functional*

$$J(\varepsilon) = \|\psi(T) - \psi_{target}\|_{L^2} + \alpha \int_0^T \varepsilon(t)^2 dt,$$

where $\alpha > 0$, ψ_{target} is a given target state and ψ is the solution of (1) *(see* [28] *and references therein for details about this problem). The critical point equations read:*

$$\begin{cases} i\partial_t \psi(x,t) = (H_0(x) - \mu(x)\varepsilon(t))\psi(x,t), \\ \psi(x,0) = \psi_0(x), \end{cases} .$$

$$\varepsilon(t) = -\frac{1}{\alpha}\langle \chi(t), \mu\psi(t)\rangle_{L^2},$$

$$\begin{cases} i\partial_t \chi(x,t) = (H_0(x) - \mu(x)\varepsilon(t))\chi(x,t), \\ \chi(x,T) = \psi_{target} - \psi(x,T), \end{cases} .$$

Using the above equation and thanks to the L^2 norm preservation (see (4)*), one finds that $|\varepsilon(t)| \leq \frac{2}{\alpha}\|\mu\|_{\mathcal{L}(H^2)}$ which means that the hypothesis $\mathcal{H}$ is satisfied with $\varepsilon_{min} = -\frac{2}{\alpha}\|\mu\|_{\mathcal{L}(H^2)}$, $\varepsilon_{max} = \frac{2}{\alpha}\|\mu\|_{\mathcal{L}(H^2)}$.*

In order to solve numerically equation (3), the "toolkit" method proceeds as follows.

Algorithm 1. *("toolkit" method)*

(1) *Preprocessing. Precompute the "toolkit", i.e. the set of propagators:*

$$S_\ell(\Delta t) \text{ for } \ell = 0, \cdots, m,$$

where $(S_\ell(t))_{t\in\mathbb{R}}$ denotes the one-parameter semi-group generated by the operator $H_0 - \mu\bar{\varepsilon}_\ell$, the sequence $(\varepsilon_\ell)_{\ell=0,\cdots,m}$, being defined by (5).

(2) *Given a control field $\varepsilon \in L^2$ satisfying $\mathcal{H}$ and $\psi_0^K = \psi_0$, the sequence $(\psi_j^K)_{j=0,\dots,N}$ that approximates $(\psi(t_j))_{j=0,\dots,N}$, is obtained recursively by iterating the following loop:*

(a) *Find:*

$$\ell_j = \text{argmin}_{\ell=1,\cdots,m}\{|\varepsilon(t_{j+\frac{1}{2}}) - \bar{\varepsilon}_\ell|\},$$

(b) *Set* $\psi^K_{j+1} = S_{\ell_j}(\Delta t)\psi^K_j$.

In this "toolkit" approximation, we consider that the changes in the Hamiltonian $H(t) := H_0 - \mu\varepsilon(t)$ can be neglected over a time step Δt. In this way, if $\Delta\varepsilon = 0$ (infinite "toolkit"), and for a relevant time discretization, the simulation corresponding to piecewise constant control fields is exact (see [29], for more details about the use of piecewise constant function in quantum control). Such a property does not hold with methods that approximate the exponential, e.g. the second order Strang operator splitting. Indeed, these approaches introduce an algebraic error, due to the non-commutation of the operators H_0 and μ that is consequently proportional to $\|\varepsilon\|_{L^\infty(0,T)}$.

Remark 2. *In the original form of the "toolkit method"* [30, 14, 15]*, the mid-point choice proposed in Step 2a of Algorithm 1 is not considered. Yet, the introduction of this strategy enables us to improve the order of the method (see the analysis hereafter).*

3.2. **Scope and numerical considerations on the "toolkit" method.** The 'toolkit' procedure relies on the precomputation of the matrices $S_\ell(\Delta t)$ for multiple values of ε_ℓ and is used in applications that require repeated resolutions of the Schrödinger equation such as control framework or inverse problems. The pre-computation will be a good investment as soon as the number of resolutions is high enough.

However, even for the control or inverse problems not all circumstances are fitted to the use of the method above. The number m of matrices $S_\ell(\Delta t)$ to be computed is not necessarily a severe limitation as this can be trivially parallelized (see also our second improvement of the method, Sec. 5, which enables to greatly reduce the toolkit size). The obvious limitation arises when the computation of $S_\ell(\Delta t)$ is difficult, for instance when the system is posed in a high spacial dimension γ. If the matrix of $H_0 - \mu\varepsilon_\ell$ in the Galerkin basis containing N_γ functions is not sparse the computation can scale as high as N_γ^3.

Even then, this scaling is routine for density matrix computations: in contrast to (1) in this formulation the evolving object is not the wavefunction but a density matrix operator which is a (self-adjoint trace-class Hilbert-Schmidt) operator on $L^2(\mathbb{R}^\gamma)$).

It is not the object of the paper to propose novel space discretization of the multi-dimensional TDSE equation neither general purpose methods for computing the exponential of a matrix [31] so we will suppose that the user will select a setting that either avoids a full matrix $H_0 - \mu\varepsilon_\ell$ or manages to obtain a small Galerkin basis, such as those arising from spectral methods or reduced basis [32, 33] (see [34] for an application in quantum chemistry). Yet another alternative is to use a low rank representation of $S_\ell(\Delta t)$ i.e. the projection of $S_\ell(\Delta t)$ on a small number of eigenvectors of $H_0 - \mu\varepsilon_\ell$; the eigenvectors can be computed without a full diagonalization of the matrix using e.g., Lanczos's method [35]. To summarize, the "toolkit" method is proposed in several contexts including

- the low dimensional systems
- any situation when a density matrix computation is possible
- the situation in (2) when the problem is not an individual system by itself but a high number of e.g., identical systems orientated differently with respect to the laser field. Note that in this case the propagators $S_\ell(\Delta t)$ are common for various values of $\xi_k \in [-1, 1]$.

3.3. **Analysis of the method.** Let us now present an error analysis of the "toolkit method". More precisely, this section aims at proving the following result:

Theorem 1. *Let $\varepsilon \in W^{2,\infty}(0,T)$ and ψ the corresponding solution of (3). Let ψ^K be the approximation of ψ obtained with Algorithm 1. There exists $\lambda_1 > 0$, $\lambda_2 > 0$, that do not depend on $\|\varepsilon\|_{L^\infty(0,T)}$ such that, for every $\Delta t, \Delta\varepsilon \leq 1$,*

$$\|\psi(T) - \psi^K(T)\|_{L^2} \leq \lambda_1 \Delta\varepsilon + \lambda_2 \Delta t^2. \tag{6}$$

Moreover, there exists $\nu_1 > 0$, $\nu_2 > 0$ depending on $\|\varepsilon\|_{W^{1,1}(0,T)}$ such that, for every $\Delta t, \Delta\varepsilon \leq 1$:

$$\|\psi(T) - \psi^K(T)\|_{H^2} \leq \nu_1 \Delta\varepsilon + \nu_2 \Delta t^2. \tag{7}$$

Remark 3. *This result shows that the "toolkit" method enables to work with large control fields, transferring the computational effort due to such cases to the preprocessing step: given $\Delta\varepsilon$, the computational cost of this step only depends on the norm $\|\varepsilon\|_{L^\infty(0,T)}$, i.e., on Hypothesis $\mathcal{H}$.*

Proof. To obtain (6) and (7), we will focus first on the local error, i.e. the approximation obtained on one time step $[t_j, t_{j+1}]$.
The sequence $(\psi_j^K)_{j=0,\dots,N}$ is a time discretization of the solution of

$$\begin{cases} i\partial_t \psi^K(t) = (H_0 - \mu\bar{\varepsilon}(t))\,\psi^K(t), & \mathbb{R}^\gamma \times (0,T) \\ \psi^K(0) = \psi_0, & \mathbb{R}^\gamma \end{cases} \tag{8}$$

where the space variable has been omitted and $\bar{\varepsilon}(t) = \bar{\varepsilon}_{\ell_j}$ is constant over each interval $[t_j, t_{j+1}[= [j\Delta t, (j+1)\Delta t[$, with $j = 0, ..., N-1$. We denote by $\big(S_j(t)\big)_{j=0,\dots,N-1}$ (instead of S_{ℓ_j}) the one-parameter semi-group generated by the operator $H_0 - \mu\bar{\varepsilon}_{\ell_j}$ and we introduce

$$\delta(t) = \varepsilon(t) - \bar{\varepsilon}$$

where $\bar{\varepsilon}$ (instead of $\bar{\varepsilon}_{\ell_j}$) is the constant value of $\bar{\varepsilon}(t)$ over $[t_j, t_{j+1}]$. Therefore, the solution ψ of (3) is actually the solution of the integral equation, settled for $t \in [t_j, t_{j+1}[$:

$$\psi(t) = S_j(t - t_j)\psi(t_j) + i\int_{t_j}^{t} S_j(t-s)\mu\delta(s)\psi(s)\; ds. \tag{9}$$

For the upcoming calculations, one should notice that we have

$$|\delta(t_{j+\frac{1}{2}})| \leq \frac{\Delta\varepsilon}{2} \tag{10}$$

and that for all $t \in [t_j, t_{j+1}]$,

$$|\delta(t)| \leq \frac{1}{2}\left(\Delta\varepsilon + \|\dot{\varepsilon}\|_{L^\infty(0,T)}\Delta t\right). \tag{11}$$

We consider the following decomposition:

$$\begin{aligned} \psi(T) - \psi^K(T) &= \psi(T) - S_{N-1}(\Delta t)\psi(t_{N-1}) \\ &+ \sum_{j=0}^{N-2} S_{N-1}(\Delta t)\dots S_{j+1}(\Delta t)\big(\psi(t_{j+1}) - S_j(\Delta t)\psi(t_j)\big) \\ &+ S_{N-1}(\Delta t)\dots S_0(\Delta t)\psi_0 - \psi^K(T) \end{aligned}$$

where the last line is equal to 0 since ψ^K satisfies (8) on $[0,T]$.

From now on and in all the following sections, we will consider either that $\|\psi_0\|_{L^2} = 1$ or that $\|\psi_0\|_{H^2} = 1$. From (4), we know that the operators S_j are isometries in L^2. Therefore, the use of a triangular inequality brings

$$\|\psi(T)-\psi^K(T)\|_{L^2} \leq \sum_{j=0}^{N-1} \|\psi(t_{j+1})-S_j(\Delta t)\psi(t_j)\|_{L^2}\,. \tag{12}$$

Given $1 \leq j \leq N-1$, we then calculate and estimate the consistency error as follows:

$$\begin{aligned}
\psi(t_{j+1})-S_j(\Delta t)\psi(t_j) &= i\int_{t_j}^{t_{j+1}} S_j(t_{j+1}-s)\delta(s)\mu\psi(s)\,ds\\
&= i\int_{t_j}^{t_{j+1}} S_j(t_{j+1}-s)\delta(s)\mu\left(\psi(s)-S_j(s-t_j)\psi(t_j)\right)\,ds\\
&\quad + i\int_{t_j}^{t_{j+1}} S_j(t_{j+1}-s)\delta(s)\mu S_j(s-t_j)\psi(t_j)\,ds.\\
&= i\int_{t_j}^{t_{j+1}} S_j(t_{j+1}-s)\delta(s)\mu\left(\psi(s)-S_j(s-t_j)\psi(t_j)\right)\,ds\\
&\quad + i\int_{t_j}^{t_{j+1}} \delta(s)\varphi_j(s)\psi(t_j)\,ds
\end{aligned} \tag{13}$$

where $\varphi_j(s) := S_j(t_{j+1}-s)\mu S_j(s-t_j) \in \mathcal{L}(L^2)$.

In what follows, we work in parallel on L^2 and H^2-estimates of $\psi(t_{j+1})-S_j(\Delta t)\psi(t_j)$. We will need basic L^2 and H^2-estimates of $\psi(t)-S_j(t-t_j)\psi(t_j)$ for the study of the first integral term of (13), the second one will be dealt with using a Taylor expansion of $\delta(t)$.

From Lemma 1, (9) and (11), it is easy to obtain coarse estimates of $\psi(t)-S_j(t-t_j)\psi(t_j)$. Indeed, for all t in $[t_j,t_{j+1}]$, one can write

$$\begin{aligned}
\|\psi(t)-S_j(t-t_j)\psi(t_j)\|_{L^2} &= \left\| i\int_{t_j}^{t} S_j(t-s)\mu\delta(s)\psi(s)\;ds\right\|_{L^2}\\
&\leq \int_{t_j}^{t_{j+1}} \|S_j(t-s)\mu\delta(s)\psi(s)\|_{L^2}\;ds\\
&\leq \Delta t\|\mu\|_{\mathcal{L}(L^2)}\frac{1}{2}\left(\Delta\varepsilon+\|\dot\varepsilon\|_{L^\infty(0,T)}\Delta t\right)\|\psi_0\|_{L^2}\\
&\leq \frac{1}{2}\|\mu\|_{\mathcal{L}(L^2)}\left(\Delta\varepsilon+\|\dot\varepsilon\|_{L^\infty(0,T)}\Delta t\right)\Delta t
\end{aligned} \tag{14}$$

and the H^2-estimate gives

$$\|\psi(t)-S_j(t-t_j)\psi(t_j)\|_{H^2} \leq K\left(1+\|\varepsilon\|_{W^{1,1}(0,T)}\|\mu\|_{\mathcal{L}(H^2)}\right)\left(\Delta\varepsilon+\|\dot\varepsilon\|_{L^\infty(0,T)}\Delta t\right)\Delta t \tag{15}$$

where $K = K(\|\mu\|_{\mathcal{L}(H^2)},\varepsilon_{\max})$ is a generic constant that estimates every $\|S_j\|_{\mathcal{L}(H^2)}$.
Therefore, we can obtain more accurate estimates of the first integral term of (13). Thanks to

(14), we obtain

$$
\begin{aligned}
&\left\| i \int_{t_j}^{t_{j+1}} S_j(t_{j+1}-s)\delta(s)\mu\left(\psi(s)-S_j(s-t_j)\psi(t_j)\right)\,ds \right\|_{L^2} \\
&\leq \frac{1}{2}\|\mu\|_{\mathcal{L}(L^2)}\left(\Delta\varepsilon + \|\dot\varepsilon\|_{L^\infty(0,T)}\Delta t\right)\Delta t \sup_{t\in[t_j,t_{j+1}]}\|\psi(s)-S_j(s-t_j)\psi(t_j)\|_{L^2} \\
&\leq \frac{1}{4}\|\mu\|^2_{\mathcal{L}(L^2)}\left(\Delta\varepsilon + \|\dot\varepsilon\|_{L^\infty(0,T)}\Delta t\right)^2\Delta t^2 \\
&\leq \frac{1}{2}\|\mu\|^2_{\mathcal{L}(L^2)}\left(\Delta\varepsilon^2 + \|\dot\varepsilon\|^2_{L^\infty(0,T)}\Delta t^2\right)\Delta t^2. \qquad (16)
\end{aligned}
$$

Working now on the H^2-estimate, we deduce from (15) in the same way that

$$
\begin{aligned}
(17)\quad &\left\| i \int_{t_j}^{t_{j+1}} S_j(t_{j+1}-s)\delta(s)\mu\left(\psi(s)-S_j(s-t_j)\psi(t_j)\right)\,ds \right\|_{H^2} \\
&\qquad \leq K\left(1+\|\mu\|_{\mathcal{L}(H^2)}\|\varepsilon\|_{W^{1,1}(0,T)}\right)\left(\Delta\varepsilon^2 + \|\dot\varepsilon\|^2_{L^\infty(0,T)}\Delta t^2\right)\Delta t^2.
\end{aligned}
$$

In the two cases (L^2 and H^2), estimates are stronger than the ones we look for, and we can focus on the second integral term of (13) we want to deal with.

We first consider

$$
\varphi_j : \begin{array}{ll} [t_j,t_{j+1}] & \to \mathcal{L}(H^2) \\ s & \mapsto S_j(t_{j+1}-s)\mu S_j(s-t_j) \end{array} \qquad (18)
$$

and note that for all $\psi \in H^2$, $\|\varphi_j(s)\psi\|_{H^2} = \|S_j(t_{j+1}-s)\mu S_j(s-t_j)\psi\|_{H^2} \leq K\|\psi\|_{H^2}$ so that

$$
\forall s \in [t_j,t_{j+1}],\ \|\varphi_j(s)\|_{\mathcal{L}(H^2)} \leq K\|\mu\|_{\mathcal{L}(H^2)}. \qquad (19)
$$

Let us now consider the derivatives of $\varphi_j(s)$. Since $(S_j(t))_{t\in\mathbb{R}}$ denotes the one-parameter semi-group generated by the operator $H_0-\mu\bar\varepsilon$, the $\mathcal{L}(H^2)$ identity

$$
\partial_t S_j(t) = -i\left(H_0-\mu\bar\varepsilon\right)S_j(t)
$$

holds and minor calculations give, $\forall s \in [t_j,t_{j+1}]$,

$$
\begin{aligned}
\partial_s\varphi_j(s) &= iS_j(t_{j+1}-s)[H_0,\mu]S_j(s-t_j) \\
\partial^2_{ss}\varphi_j(s) &= S_j(t_{j+1}-s)\big[[H_0,\mu],H_0-\mu\bar\varepsilon\big]S_j(s-t_j).
\end{aligned}
$$

Therefore,

$$
\begin{aligned}
\|\partial_s\varphi_j(s)\|_{\mathcal{L}(H^2)} &\leq K\|[H_0,\mu]\|_{\mathcal{L}(H^2)} \\
\|\partial^2_{ss}\varphi_j(s)\|_{\mathcal{L}(H^2)} &\leq K\left\|\big[[H_0,\mu],H_0-\mu\bar\varepsilon\big]\right\|_{\mathcal{L}(H^2)}.
\end{aligned} \qquad (20)
$$

If we consider the L^2-analysis of the method, then $\varphi_j(s) \in \mathcal{L}(L^2)$ and $\forall s \in [t_j,t_{j+1}]$,

$$
\begin{aligned}
\|\varphi_j(s)\|_{\mathcal{L}(L^2)} &\leq \|\mu\|_{\mathcal{L}(L^2)} \\
\|\partial_s\varphi_j(s)\|_{\mathcal{L}(L^2)} &\leq \|[H_0,\mu]\|_{\mathcal{L}(L^2)} \\
\|\partial^2_{ss}\varphi_j(s)\|_{\mathcal{L}(L^2)} &\leq \left\|\big[[H_0,\mu],H_0-\mu\bar\varepsilon\big]\right\|_{\mathcal{L}(L^2)}.
\end{aligned} \qquad (21)
$$

Let us now write the third order Taylor expansion of $t \mapsto \delta(t) = \varepsilon(t) - \bar{\varepsilon}$ in a neighborhood of $t_{j+\frac{1}{2}}$:

$$\begin{aligned}\delta(s) &= \delta(t_{j+\frac{1}{2}}) + (s - t_{j+\frac{1}{2}})\dot{\delta}(t_{j+\frac{1}{2}}) + \frac{1}{2}(s - t_{j+\frac{1}{2}})^2\ddot{\delta}(\theta(s)) \\ &= \delta(t_{j+\frac{1}{2}}) + (s - t_{j+\frac{1}{2}})\dot{\varepsilon}(t_{j+\frac{1}{2}}) + \frac{1}{2}(s - t_{j+\frac{1}{2}})^2\ddot{\varepsilon}\big(\theta(s)\big),\end{aligned}$$

with $\theta(s) \in [t_j, t_{j+1}]$. We now focus on estimating the term $i\displaystyle\int_{t_j}^{t_{j+1}} \delta(s)\varphi_j(s)\psi(t_j)\,ds$. By means of (21) and the L^2-norm conservation, we obtain

$$\left\| \int_{t_j}^{t_{j+1}} \delta(t_{j+\frac{1}{2}})\varphi_j(s)\psi(t_j)ds \right\|_{L^2} \leq \frac{1}{2}\|\mu\|_{\mathcal{L}(L^2)}\Delta\varepsilon\Delta t,$$

$$\begin{aligned}&\left\| \int_{t_j}^{t_{j+1}} \left(s - t_{j+\frac{1}{2}}\right)\dot{\varepsilon}\left(t_{j+\frac{1}{2}}\right)\varphi_j(s)\psi(t_j)ds \right\|_{L^2} \\ &= \left\| \dot{\varepsilon}\left(t_{j+\frac{1}{2}}\right)\int_0^{\frac{1}{2}\Delta t} s\,\left(\varphi(t_{j+\frac{1}{2}} + s) - \varphi(t_{j+\frac{1}{2}} - s)\right)\psi(t_j)ds \right\|_{L^2} \\ &= \left\| \dot{\varepsilon}\left(t_{j+\frac{1}{2}}\right)\int_0^{\frac{1}{2}\Delta t}\int_{t_{j+\frac{1}{2}}-s}^{t_{j+\frac{1}{2}}+s} s\partial_u\varphi(u)\psi(t_j)\;duds \right\|_{L^2} \\ &\leq \frac{1}{12}\|[H_0, \mu]\|_{\mathcal{L}(L^2)}\|\dot{\varepsilon}\|_{L^\infty(t_j, t_{j+1})}\Delta t^3\end{aligned}$$

and

$$\left\| \int_{t_j}^{t_{j+1}} \frac{1}{2}\left(s - t_{j+\frac{1}{2}}\right)^2\ddot{\varepsilon}(\theta(s))\varphi_j(s)\psi(t_j)ds \right\|_{L^2} \leq \frac{1}{24}\|\mu\|_{\mathcal{L}(L^2)}\|\ddot{\varepsilon}\|_{L^\infty(t_j, t_{j+1})}\Delta t^3.$$

Combining these results with (16), we estimate (13) as follows:

$$\begin{aligned}\|\psi(t_{j+1}) - S_j(\Delta t)\psi(t_j)\|_{L^2} \leq\;& \frac{1}{2}\|\mu\|^2_{\mathcal{L}(L^2)}\left(\Delta\varepsilon^2\Delta t^2 + \|\dot{\varepsilon}\|^2_\infty\Delta t^4\right) \\ &+ \frac{1}{2}\|\mu\|_{\mathcal{L}(L^2)}\Delta\varepsilon\Delta t \\ &+ \frac{1}{24}\left(2\|[H_0, \mu]\|_{\mathcal{L}(L^2)}\|\dot{\varepsilon}\|_\infty + \|\mu\|_{\mathcal{L}(L^2)}\|\ddot{\varepsilon}\|_\infty\right)\Delta t^3,\end{aligned}$$

with $\|\cdot\|_{L^\infty(0,T)} = \|\cdot\|_\infty$. By means of (12), the global L^2-estimate is then:

$$\begin{aligned}\|\psi(T) - \psi^K(T)\|_{L^2} &\leq \sum_{j=0}^{N-1}\|\psi(t_{j+1}) - S_j(\Delta t)\psi(t_j)\|_{L^2} \\ &\leq \frac{T}{2}\|\mu\|^2_{\mathcal{L}(L^2)}\left(\Delta\varepsilon^2\Delta t + \|\dot{\varepsilon}\|^2_\infty\Delta t^3\right) + \frac{T}{2}\|\mu\|_{\mathcal{L}(L^2)}\Delta\varepsilon \\ &\quad + \frac{T}{24}\left(2\|[H_0, \mu]\|_{\mathcal{L}(L^2)}\|\dot{\varepsilon}\|_\infty + \|\mu\|_{\mathcal{L}(L^2)}\|\ddot{\varepsilon}\|_\infty\right)\Delta t^2,\end{aligned}$$

and (6) can be deduced with the following constants λ_1 and λ_2 independent of $\|\varepsilon\|_{L^\infty(0,T)}$ (reminding the reader that we assume $\Delta\varepsilon < 1$ and $\Delta t < 1$):

$$\begin{aligned}\lambda_1 &= \frac{T}{2}\|\mu\|_{\mathcal{L}(L^2)}\left(1+\|\mu\|_{\mathcal{L}(L^2)}\right),\\ \lambda_2 &= \frac{T}{2}\,\|\mu\|^2_{\mathcal{L}(L^2)}\|\dot\varepsilon\|^2_\infty + \frac{T}{12}\|[H_0,\mu]\|_{\mathcal{L}(L^2)}\|\dot\varepsilon\|_\infty + \frac{T}{24}\|\mu\|_{\mathcal{L}(L^2)}\|\ddot\varepsilon\|_\infty.\end{aligned}$$

Let us now prove the H^2 estimate. By means of (17), (19) and (20) and keeping in mind that K is a generic constant depending on $\|\mu\|_{\mathcal{L}(H^2)}$ and $\varepsilon_{\max}$, we can repeat the previous analysis to find the local estimate:

$$\begin{aligned}\|\psi(t_{j+1}) - S_j(\Delta t)\psi(t_j)\|_{H^2} \quad &\leq \quad K\left(1+\|\mu\|_{\mathcal{L}(H^2)}\|\varepsilon\|_{W^{1,1}(0,T)}\right)\Big(\Delta\varepsilon^2\Delta t^2 + \|\dot\varepsilon\|^2_{L^\infty(0,T)}\Delta t^4\Big)\\ &\quad + K\Delta\varepsilon\Delta t + K\Big(\|[H_0,\mu]\|_{\mathcal{L}(H^2)}\|\dot\varepsilon\|_{L^\infty} + \|\ddot\varepsilon\|_{L^\infty}\Big)\Delta t^3.\end{aligned}$$

Since one can prove that we can actually write a more precise estimate of $S_j(\Delta t)$ and replace K by $1 + C\Delta t$ (see properties (4)), we get:

$$\|S_j(\Delta t)\|_{\mathcal{L}(H^2)} \leq 1 + C\Delta t$$

and since we have the following intermediate result, where $M > 0$ depends on $\|\mu\|_{\mathcal{L}(H^2)}$, $\varepsilon_{\max}$ and T but is independent of N:

$$\sum_{j=0}^{N-1}(1+C\Delta t)^{N-j}\Delta t \leq M.$$

The global estimate is obtained as follows:

$$\begin{aligned}\|\psi(T)-\psi^K(T)\|_{H^2} \quad &\leq \quad \sum_{j=0}^{N-1} K^{N-j-1}\|\psi(t_{j+1}) - S_j(\Delta t)\psi(t_j)\|_{H^2}\\ &\leq \quad \sum_{j=0}^{N-1}(1+C\Delta t)^{N-j}(1+\|\varepsilon\|_{W^{1,1}(0,T)})\left(\Delta\varepsilon^2\Delta t^2 + \|\dot\varepsilon\|^2_\infty\Delta t^4\right)\\ &\quad + \sum_{j=0}^{N-1}(1+C\Delta t)^{N-j}\left(\Delta\varepsilon\Delta t + \left(\|[H_0,\mu]\|_{\mathcal{L}(H^2)}\|\dot\varepsilon\|_\infty + \|\ddot\varepsilon\|_\infty\right)\Delta t^3\right)\\ &\leq \quad M(1+\|\varepsilon\|_{W^{1,1}(0,T)})\left(\Delta\varepsilon^2\Delta t + \|\dot\varepsilon\|^2_\infty\Delta t^3\right) + M\Delta\varepsilon\\ &\quad + M\left(\|[H_0,\mu]\|_{\mathcal{L}(H^2)}\|\dot\varepsilon\|_\infty + \|\ddot\varepsilon\|_\infty\right)\Delta t^2.\end{aligned}$$

We finally get ν_1 and ν_2 (assuming $\Delta\varepsilon < 1$ and $\Delta t < 1$) and conclude the proof of Theorem 1:

$$\begin{aligned}\nu_1 &= M(1+T+\|\varepsilon\|_{W^{1,1}(0,T)}T)\\ \nu_2 &= M\left((1+\|\varepsilon\|_{W^{1,1}(0,T)})\|\dot\varepsilon\|^2_\infty + \|[H_0,\mu]\|_{\mathcal{L}(H^2)}\|\dot\varepsilon\|_\infty + \|\ddot\varepsilon\|_\infty\right).\end{aligned}$$

□

Remark 4. *The estimate* (6) *is consistent with the fact that Algorithm 1 used with a relevant time discretization is exact for the piecewise constant control fields.*

4. Improvement in the limit of low intensities

We now describe a way to improve the time order of the previous algorithm. Since some constants in the following analysis depend in this case of the L^∞-norm of the field and the method requires that the "toolkit" size scales $\Delta t^3(\varepsilon_{\max} - \varepsilon_{\min})$, it applies in the case of (L^∞-) small control fields.

4.1. **Algorithm.** The algorithm we propose mixes the "toolkit" and the splitting approaches, in the sense that it applies sequentially various operators to correct the third order local error that appears in the proof of Theorem 1.

Algorithm 2. *(Improved "toolkit" method for low intensities)*

(1) *Preprocessing. Precompute the "toolkit", i.e. the set of propagators:*

$$S_\ell(\Delta t) \text{ for } \ell = 0, \cdots, m,$$

where $(S_\ell(t))_{t\in\mathbb{R}}$ *denotes the one-parameter group generated by the operator* $H_0 - \mu\bar{\varepsilon}_\ell$, *the sequence* $(\varepsilon_\ell)_{\ell=0,\cdots,m}$, *being defined by* (5). *Include in this set the two special elements:*

$$\Omega = e^{\frac{1}{12}[H_0,\mu]\Delta t^3}, \Theta = e^{\frac{i}{24}\mu\Delta t^3}$$

and the initial exponents α_0 *and* β_0 *such that* (ε *being extended as an even function on [-T,0]):*

$$\begin{aligned}\alpha_0 &:= \frac{\varepsilon(\Delta t) - \varepsilon(0)}{\Delta t} = \dot{\varepsilon}(t_{\frac{1}{2}}) + \mathcal{O}(\Delta t^2),\\ \beta_0 &:= \frac{\varepsilon(t_1) - 2\varepsilon(t_{\frac{1}{2}}) + \varepsilon(0)}{\Delta t^2} = \ddot{\varepsilon}(t_{\frac{1}{2}}) + \mathcal{O}(\Delta t^2).\end{aligned}$$

(2) *Given a control field* $\varepsilon \in L^\infty$ *satisfying* $\mathcal{H}$ *and* $\psi_0^{IK} = \Omega^{\alpha_0}\Theta^{\beta_0}\psi_0$, *the sequence* $(\psi_j^{IK})_{j=0,\dots,N}$ *that approximates* $(\psi(t_j))_{j=0,\dots,N}$, *is obtained recursively by iterating the following loop:*

(a) *Find:*

$$\ell_j = \operatorname{argmin}_{\ell=1,\cdots,m}\{|\varepsilon(t_{j+1/2}) - \bar{\varepsilon}_\ell|\},$$

(b) *Compute* α_j *and* β_j *such that:*

$$\alpha_j := \frac{\varepsilon(t_{j+1}) - \varepsilon(t_j)}{\Delta t} = \dot{\varepsilon}(t_{j+\frac{1}{2}}) + \mathcal{O}(\Delta t^2), \tag{22}$$

$$\beta_j := \frac{\varepsilon(t_{j+1}) - 2\varepsilon(t_{j+\frac{1}{2}}) + \varepsilon(t_j)}{\Delta t^2} = \ddot{\varepsilon}(t_{j+\frac{1}{2}}) + \mathcal{O}(\Delta t^2). \tag{23}$$

(c) *Set* $\psi_{j+1}^{IK} = S_{\ell_j}(\Delta t)\Omega^{\alpha_j}\Theta^{\beta_j}\psi_j^{IK}$.

In many cases, e.g. in the experimental frameworks, only the values of the field can be handled. The use of exact values for the time derivatives has then to be avoided when possible. This motivates the introduction of approximations (22) and (23) of $\dot{\varepsilon}(t_j)$ and $\ddot{\varepsilon}(t_j)$ in the latest definitions. The analysis presented hereafter shows that this does not deteriorate the order of the method.

In this method, one must perform two online matrices exponentiations. By working in a basis where one of these two matrices is diagonal, the cost of Step *2c* can be reduced to one exponentiation, making the cost of this method equivalent the second order Strang operator splitting.

4.2. **Analysis of the method.** We can now repeat the analysis that has been done in the proof of Theorem 1 to obtain the following estimate.

Theorem 2. *Let* $\varepsilon \in W^{2,\infty}(0,T)$, ψ *be the corresponding solution of* (3) *and* ψ^{IK} *the approximation of* ψ *obtained with Algorithm 2. There exists* $\lambda'_1 > 0$, $\lambda'_2 > 0$, *with* λ'_1 *independent of* $\|\varepsilon\|_{L^\infty(0,T)}$ *such that, for every* $\Delta t, \Delta\varepsilon \leq 1$,

$$\|\psi(T) - \psi^{IK}(T)\|_{L^2} \leq \lambda'_1 \Delta\varepsilon + \lambda'_2 \Delta t^3.$$

Proof. In the framework of this new algorithm, we note that on every time interval $]t_j, t_{j+1}[$, the approximation ψ^{IK} is the solution of the evolution equation:

$$\left\{ \begin{array}{lll} i\partial_t \psi^{IK}(t) & = (H_0 - \mu\bar{\varepsilon})\, \psi^{IK}(t), & \mathbb{R}^\gamma \times (t_j, t_{j+1}) \\ \psi^{IK}(t_j^+) & = \Omega^{\alpha_j}\Theta^{\beta_j}\psi^{IK}(t_j^-) & \mathbb{R}^\gamma \end{array} \right. \tag{24}$$

where we set $\psi(0^-) = \psi_0$. We will keep the notations (S_j, $\delta(t)$, φ,...) of the proof of Theorem 1, and we first focus on the local error analysis. We consider the following decomposition:

$$\begin{aligned} \psi(T) - \psi^{IK}(T) &= \psi(T) - S_{N-1}(\Delta t)\Omega^{\alpha_{N-1}}\Theta^{\beta_{N-1}}\psi(t_{N-1}) \\ &+ \sum_{j=0}^{N-2} S_{N-1}(\Delta t)\Omega^{\alpha_{N-1}}\Theta^{\beta_{N-1}} \ldots S_{j+1}(\Delta t)\Omega^{\alpha_{j+1}}\Theta^{\beta_{j+1}} \\ &\qquad \times \big(\psi(t_{j+1}) - S_j(\Delta t)\Omega^{\alpha_j}\Theta^{\beta_j}\psi(t_j)\big) \\ &+ S_{N-1}(\Delta t)\Omega^{\alpha_{N-1}}\Theta^{\beta_{N-1}} \ldots S_0(\Delta t)\Omega^{\alpha_0}\Theta^{\beta_0}\psi_0 - \psi^{IK}(T) \end{aligned}$$

where the last line is equal to 0 since ψ^{IK} satisfies (24) on $[0,T]$.
The operators S_j are isometries in L^2, we will consider that $\|\psi_0\|_{L^2} = 1$ and we also have, for all j

$$\Omega^{\alpha_j}\Theta^{\beta_j} = e^{\frac{\alpha_j}{12}[H_0,\mu]\Delta t^3} e^{\frac{i\beta_j}{24}\mu\Delta t^3} = \mathrm{Id} + \left(\frac{\alpha_j}{12}[H_0,\mu] + \frac{i\beta_j}{24}\mu\right)\Delta t^3 + \mathrm{Id}\ \mathcal{O}(\Delta t^6) \tag{25}$$

and thus

$$\left\|\Omega^{\alpha_j}\Theta^{\beta_j}\right\|_{\mathcal{L}(L^2)} \leq 1 + \mathcal{O}(\Delta t^3). \tag{26}$$

Therefore, the use of a triangular inequality brings

$$\|\psi(T) - \psi^{IK}(T)\|_{L^2} \leq (1 + \mathcal{O}(\Delta t^2)) \sum_{j=0}^{N-1} \left\|\psi(t_{j+1}) - S_j(\Delta t)\Omega^{\alpha_j}\Theta^{\beta_j}\psi(t_j)\right\|_{L^2} \tag{27}$$

and we will calculate and estimate in L^2-norm for all j the difference

$$\begin{aligned} & \psi(t_{j+1}) - S_j(\Delta t)\Omega^{\alpha_j}\Theta^{\beta_j}\psi(t_j) \\ = \ & S_j(\Delta t)\psi(t_j) + i\int_{t_j}^{t_{j+1}} S_j(t_{j+1}-s)\delta(s)\mu\psi(s)\,ds - S_j(\Delta t)\Omega^{\alpha_j}\Theta^{\beta_j}\psi(t_j) \\ = \ & S_j(\Delta t)\left(\mathrm{Id} - \Omega^{\alpha_j}\Theta^{\beta_j}\right)\psi(t_j) + i\int_{t_j}^{t_{j+1}} S_j(t_{j+1}-s)\delta(s)\mu\psi(s)\,ds. \end{aligned}$$

We define $Y(s) = \psi(s) - S_j(s-t_j)\Omega^{\alpha_j}\Theta^{\beta_j}\psi(t_j)$ for all $s \in [t_j, t_{j+1}]$ and obtain

$$Y(t_{j+1}) = S_j(\Delta t)\left(\mathrm{Id} - \Omega^{\alpha_j}\Theta^{\beta_j}\right)\psi(t_j) + i\int_{t_j}^{t_{j+1}} S_j(t_{j+1}-s)\delta(s)\mu Y(s)\,ds + i\int_{t_j}^{t_{j+1}} \delta(s)\varphi_j(s)\Omega^{\alpha_j}\Theta^{\beta_j}\psi(t_j)\,ds \tag{28}$$

where $\varphi_j(s) := S_j(t_{j+1}-s)\mu S_j(s-t_j)$ and its derivatives have been estimated in L^2 in (21). As we did in Theorem 1, we start with an estimate of the first integral term of (28). For all $t \in]t_j, t_{j+1}]$, we can write:

$$\begin{aligned} Y(t) &= \psi(t) - S_j(t-t_j)\psi(t_j) + S_j(t-t_j)\psi(t_j) - S_j(t-t_j)\Omega^{\alpha_j}\Theta^{\beta_j}\psi(t_j) \\ &= \int_{t_j}^{t} S_j(t-s)\delta(s)\mu\psi(s)\ ds + S_j(t-t_j)\left(\mathrm{Id} - \Omega^{\alpha_j}\Theta^{\beta_j}\right)\psi(t_j). \end{aligned}$$

Moreover, for all $t \in]t_j, t_{j+1}]$, we have

$$\|Y(t)\|_{L^2} \leq \left\| \int_{t_j}^{t} S_j(t-s)\delta(s)\mu\psi(s)\ ds \right\|_{L^2} + \left\| S_j(t-t_j)\left(\mathrm{Id} - \Omega^{\alpha_j}\Theta^{\beta_j}\right)\psi(t_j)\right\|_{L^2}$$

The operators S_j are isometries in L^2 and $\forall t \in [0,T]$, $\|\psi(t)\|_{L^2} = \|\psi_0\|_{L^2}$. Therefore, we deduce from (25) that

$$\left\| S_j(t-t_j)\left(\mathrm{Id} - \Omega^{\alpha_j}\Theta^{\beta_j}\right)\psi(t_j)\right\|_{L^2} \leq \left(\frac{\alpha_j}{12}\|[H_0,\mu]\|_{\mathcal{L}(L^2)} + \frac{i\beta_j}{24}\|\mu\|_{\mathcal{L}(L^2)}\right)\Delta t^3 + \mathcal{O}(\Delta t^6).$$

Since it is clear that we also have

$$\left\| \int_{t_j}^{t} S_j(t-s)\delta(s)\mu\psi(s)\ ds \right\|_{L^2} \leq \frac{1}{2}\|\mu\|_{\mathcal{L}(L^2)}\left(\Delta\varepsilon + \|\dot{\varepsilon}\|_{L^\infty(0,T)}\Delta t\right)\Delta t,$$

one can finally deduce that:

$$\begin{aligned} &\left\| i\int_{t_j}^{t_{j+1}} S_j(t_{j+1}-s)\delta(s)\mu Y(s)\ ds \right\|_{L^2} \\ &\leq \frac{1}{2}\|\mu\|_{\mathcal{L}(L^2)}\left(\Delta\varepsilon + \|\dot{\varepsilon}\|_{L^\infty(0,T)}\Delta t\right)\Delta t \sup_{t\in[t_j,t_{j+1}]}\|Y(t)\|_{L^2} \\ &\leq \frac{1}{4}\|\mu\|^2_{\mathcal{L}(L^2)}\left(\Delta\varepsilon + \|\dot{\varepsilon}\|_{L^\infty(0,T)}\Delta t\right)^2\Delta t^2 + \mathcal{O}(\Delta\varepsilon\Delta t^4) + \mathcal{O}(\Delta t^5). \end{aligned} \tag{29}$$

We focus now on the first and third terms of (28). Using (25), we get

$$S_j(\Delta t)\left(\mathrm{Id} - \Omega^{\alpha_j}\Theta^{\beta_j}\right)\psi(t_j) = -S_j(\Delta t)\left(\frac{\alpha_j}{12}[H_0,\mu] + \frac{i\beta_j}{24}\mu\right)\psi(t_j)\Delta t^3 + \psi(t_j)\mathcal{O}(\Delta t^6).$$

Let us then consider the second integral term of (28). On the one hand, we consider the fourth order expansion of $\delta = \varepsilon - \bar{\varepsilon}$ in a neighborhood of $t_{j+\frac{1}{2}}$:

$$\begin{aligned} \delta(s) &= \delta(t_{j+\frac{1}{2}}) + (s-t_{j+\frac{1}{2}})\dot{\delta}(t_{j+\frac{1}{2}}) + \frac{1}{2}(s-t_{j+\frac{1}{2}})^2\ddot{\delta}(t_{j+\frac{1}{2}}) + \frac{1}{6}(s-t_{j+\frac{1}{2}})^3\delta^{(3)}(\theta(s)) \\ &= \delta(t_{j+\frac{1}{2}}) + (s-t_{j+\frac{1}{2}})\dot{\varepsilon}(t_{j+\frac{1}{2}}) + \frac{1}{2}(s-t_{j+\frac{1}{2}})^2\ddot{\varepsilon}(t_{j+\frac{1}{2}}) + \frac{1}{6}(s-t_{j+\frac{1}{2}})^3\varepsilon^{(3)}\left(\theta(s)\right) \end{aligned}$$

where $\theta(s) \in [t_j, t_{j+1}]$. On the other hand, we calculate and/or estimate the four corresponding terms in

$$i\int_{t_j}^{t_{j+1}} \delta(s)\varphi_j(s)\Omega^{\alpha_j}\Theta^{\beta_j}\psi(t_j)\,ds.$$

From (10) and (21), the term of order 0 gives:

$$\left\| i\int_{t_j}^{t_{j+1}} \delta(t_{j+\frac{1}{2}})\varphi_j(s)\Omega^{\alpha_j}\Theta^{\beta_j}\psi(t_j)ds\right\|_{L^2} \leq \frac{1}{2}\|\mu\|^2_{\mathcal{L}(L^2)}\Delta\varepsilon\Delta t.$$

For the term of order 1, we can write

$$\begin{aligned}
& i\int_{t_j}^{t_{j+1}} (s-t_{j+\frac{1}{2}})\dot{\varepsilon}(t_{j+\frac{1}{2}})\varphi_j(s)\Omega^{\alpha_j}\Theta^{\beta_j}\psi(t_j)\ ds \\
= & \ i\ \dot{\varepsilon}\left(t_{j+\frac{1}{2}}\right)\int_0^{\frac{1}{2}\Delta t} s\big(\varphi_j(t_{j+\frac{1}{2}}+s)-\varphi_j(t_{j+\frac{1}{2}}-s)\big)\Omega^{\alpha_j}\Theta^{\beta_j}\psi(t_j)\ ds \\
= & \ i\ \dot{\varepsilon}\left(t_{j+\frac{1}{2}}\right)\int_0^{\frac{1}{2}\Delta t}\int_{t_{j+\frac{1}{2}}-s}^{t_{j+\frac{1}{2}}+s} s\partial_u\varphi_j(u)\Omega^{\alpha_j}\Theta^{\beta_j}\psi(t_j)\ duds \\
= & \ i\ \dot{\varepsilon}(t_{j+\frac{1}{2}})\int_0^{\frac{1}{2}\Delta t}\int_{t_{j+\frac{1}{2}}-s}^{t_{j+\frac{1}{2}}+s} s\big(\partial_u\varphi_j(t_j)+(u-t_j)\tau(u)\big)\Omega^{\alpha_j}\Theta^{\beta_j}\psi(t_j)\ duds \\
= & \ \frac{\dot{\varepsilon}(t_{j+\frac{1}{2}})}{12}S_j\left(\Delta t\right)[H_0,\mu]\Omega^{\alpha_j}\Theta^{\beta_j}\psi(t_j)\Delta t^3 \\
& + i\ \dot{\varepsilon}(t_{j+\frac{1}{2}})\int_0^{\frac{1}{2}\Delta t}\int_{t_{j+\frac{1}{2}}-s}^{t_{j+\frac{1}{2}}+s} s(u-t_j)\tau(u-t_j)\Omega^{\alpha_j}\Theta^{\beta_j}\psi(t_j)\ duds \\
= & \ \frac{\alpha_j}{12}S_j\left(\Delta t\right)[H_0,\mu]\psi(t_j)\Delta t^3 + S_j\left(\Delta t\right)[H_0,\mu]\psi(t_j)\mathcal{O}(\Delta t^6) \\
& + i\ \dot{\varepsilon}(t_{j+\frac{1}{2}})\int_0^{\frac{1}{2}\Delta t}\int_{t_{j+\frac{1}{2}}-s}^{t_{j+\frac{1}{2}}+s} s(u-t_j)\tau(u-t_j)\Omega^{\alpha_j}\Theta^{\beta_j}\psi(t_j)\ duds
\end{aligned}$$

where we used (22), (25) and (21) and the function $\tau: s\in[0,\Delta t]\mapsto\tau(s)\in\mathcal{L}(L^2)$ is defined as the function that appears in the following expansion of $\partial_u\varphi_j$ around t_j, for any $\psi\in L^2$

$$\begin{aligned}
\partial_u\varphi_j(u)\psi &= \partial_u\varphi_j(t_j)\psi+(u-t_j)\tau(u-t_j)\psi \\
&= iS_j(\Delta t)[H_0,\mu]\psi+(u-t_j)\tau(u-t_j)\psi.
\end{aligned}$$

Using the estimate (coming from (21))

$$\|\tau(s)\|_{\mathcal{L}(L^2)} \leq \left\|\Big[[H_0,\mu],H_0-\mu\bar{\varepsilon}\Big]\right\|_{\mathcal{L}(L^2)}\ \forall s\in[0,\Delta t], \tag{30}$$

along with $\|\psi(t_j)\|_{L^2}=\|\psi_0\|_{L^2}=1$, (22) and (26) we find that for all j,

$$\begin{aligned}
& \left\| i\dot{\varepsilon}(t_{j+\frac{1}{2}})\int_0^{\frac{1}{2}\Delta t}\int_{t_{j+\frac{1}{2}}-s}^{t_{j+\frac{1}{2}}+s} s(u-t_j)\tau(u-t_j)\Omega^{\alpha_j}\Theta^{\beta_j}\psi(t_j)\ duds\right\|_{L^2} \\
\leq & \ \left(\alpha_j+\mathcal{O}\left(\Delta t^2\right)\right)\int_0^{\frac{1}{2}\Delta t}\int_{t_{j+\frac{1}{2}}-s}^{t_{j+\frac{1}{2}}+s} s(u-t_j)\left\|\Big[[H_0,\mu],H_0-\mu\bar{\varepsilon}\Big]\right\|_{\mathcal{L}(L^2)}\|\psi(t_j)\|_{L^2}\ duds. \\
\leq & \ \frac{\alpha_j}{24}\left\|\Big[[H_0,\mu],H_0-\mu\bar{\varepsilon}\Big]\right\|_{\mathcal{L}(L^2)}\Delta t^4+\mathcal{O}\left(\Delta t^5\right).
\end{aligned}$$

We also prove easily that for all j,

$$\left\|S_j\left(\Delta t\right)[H_0,\mu]\psi(t_j)\mathcal{O}(\Delta t^6)\right\|_{L^2}=\mathcal{O}\left(\Delta t^6\right).$$

For the term of order 2, using (23), (25) and (21) and the first order expansion of φ_j around t_j, $\varphi_j(s)\psi = \varphi_j(t_j)\psi + (s-t_j)\theta(s-t_j)\psi$ for all $\psi \in L^2$, defining $\theta : s \in [0,\Delta t] \mapsto \theta(s) \in \mathcal{L}(L^2)$, we can write

$$\begin{aligned}
& i\int_{t_j}^{t_{j+1}} \frac{1}{2}(s-t_{j+\frac{1}{2}})^2 \ddot{\varepsilon}(t_{j+\frac{1}{2}})\varphi_j(s)\Omega^{\alpha_j}\Theta^{\beta_j}\psi(t_j)\ ds \\
= \ & i\ddot{\varepsilon}(t_{j+\frac{1}{2}}) \int_{t_j}^{t_{j+1}} \frac{1}{2}(s-t_{j+\frac{1}{2}})^2 \left(\varphi_j(t_j) + (s-t_j)\theta(s-t_j)\right)\Omega^{\alpha_j}\Theta^{\beta_j}\psi(t_j)\ ds \\
= \ & \frac{i\ddot{\varepsilon}(t_{j+\frac{1}{2}})}{24} S_j\left(\Delta t\right)\mu\Omega^{\alpha_j}\Theta^{\beta_j}\psi(t_j)\Delta t^3 \\
& + \frac{i\ddot{\varepsilon}(t_{j+\frac{1}{2}})}{2}\int_{t_j}^{t_{j+1}} (s-t_{j+\frac{1}{2}})^2(s-t_j)\theta(s-t_j)\Omega^{\alpha_j}\Theta^{\beta_j}\psi(t_j)\ ds \\
= \ & \frac{i\beta_j}{24} S_j\left(\Delta t\right)\mu\psi(t_j)\Delta t^3 + S_j\left(\Delta t\right)\mu\psi(t_j)\mathcal{O}(\Delta t^6) \\
& + \frac{i\ddot{\varepsilon}(t_{j+\frac{1}{2}})}{2}\int_{t_j}^{t_{j+1}} (s-t_{j+\frac{1}{2}})^2(s-t_j)\theta(s-t_j)\Omega^{\alpha_j}\Theta^{\beta_j}\psi(t_j)\ ds.
\end{aligned}$$

Using (21), we get the estimate $\|\theta(s)\|_{\mathcal{L}(L^2)} \leq \left\|\left[H_0,\mu\right]\right\|_{\mathcal{L}(L^2)}$, $\forall s \in [0,\Delta t]$, and using it with (23) and (26), we obtain that for all j,

$$\begin{aligned}
& \left\|\frac{i\ddot{\varepsilon}(t_{j+\frac{1}{2}})}{2}\int_{t_j}^{t_{j+1}} (s-t_{j+\frac{1}{2}})^2(s-t_j)\theta(s-t_j)\Omega^{\alpha_j}\Theta^{\beta_j}\psi(t_j)\ ds\right\|_{L^2} \\
\leq \ & \frac{1}{2}\left(\beta_j + \mathcal{O}\left(\Delta t^2\right)\right)\int_{t_j}^{t_{j+1}} (s-t_{j+\frac{1}{2}})^2(t_j-s)\left\|[H_0,\mu]\right\|_{\mathcal{L}(L^2)}\left\|\psi(t_j)\right\|_{L^2}\ ds \\
\leq \ & \frac{1}{2}\left(\beta_j + \mathcal{O}\left(\Delta t^2\right)\right)\left\|[H_0,\mu]\right\|_{\mathcal{L}(L^2)}\int_{-\frac{\Delta t}{2}}^{\frac{\Delta t}{2}} u^2\left(\frac{\Delta t}{2} - u\right)\ du \\
\leq \ & \frac{\beta_j}{48}\left\|[H_0,\mu]\right\|_{\mathcal{L}(L^2)}\Delta t^4 + \mathcal{O}\left(\Delta t^5\right).
\end{aligned}$$

and we also prove easily that $\left\|S_j\left(\Delta t\right)\mu\psi(t_j)\mathcal{O}(\Delta t^6)\right\|_{L^2} = \mathcal{O}\left(\Delta t^6\right).$

Combining these results with (29) into equation (28), we obtain:

$$\begin{aligned}
\|Y(t_{j+1})\|_{L^2} = \ & \left\|\psi(t_{j+1}) - S_j(\Delta t)\Omega^{\alpha_j}\Theta^{\beta_j}\psi(t_j)\right\|_{L^2} \\
\leq \ & \left\|S_j(\Delta t)\left(\mathrm{Id} - \Omega^{\alpha_j}\Theta^{\beta_j}\right)\psi(t_j) + i\int_{t_j}^{t_{j+1}}\delta(s)\varphi_j(s)\Omega^{\alpha_j}\Theta^{\beta_j}\psi(t_j)\,ds\right\|_{L^2} \\
& + \left\|i\int_{t_j}^{t_{j+1}} S_j(t_{j+1}-s)\delta(s)\mu Y(s)\,ds\right\|_{L^2} \\
\leq \ & \frac{1}{2}\|\mu\|^2_{\mathcal{L}(L^2)}\Delta\varepsilon\Delta t + \frac{\alpha_j}{24}\left\|\left[[H_0,\mu], H_0 - \mu\bar{\varepsilon}\right]\right\|_{\mathcal{L}(L^2)}\Delta t^4 \\
& + \frac{\beta_j}{48}\left\|[H_0,\mu]\right\|_{\mathcal{L}(L^2)}\Delta t^4 + \frac{1}{2}\|\mu\|^2_{\mathcal{L}(L^2)}\left(\Delta\varepsilon^2\Delta t^2 + \|\dot{\varepsilon}\|^2_{L^\infty(0,T)}\Delta t^4\right) \\
& + \mathcal{O}(\Delta\varepsilon\Delta t^4) + \mathcal{O}(\Delta t^5)
\end{aligned}$$

We have now a local in time estimate that should be traduced in a global one, and from (27), we get

$$\begin{aligned}
&\|\psi(T)-\psi^{IK}(T)\|_{L^2}\\
\leq\;& (1+\mathcal{O}(\Delta t^2))\sum_{j=0}^{N-1}\left\|\psi(t_{j+1})-S_j(\Delta t)\Omega^{\alpha_j}\Theta^{\beta_j}\psi(t_j)\right\|_{L^2}\\
\leq\;& \frac{T}{2}\|\mu\|^2_{\mathcal{L}(L^2)}\Delta\varepsilon+\frac{\alpha_j T}{24}\left\|\Big[[H_0,\mu],H_0-\mu\bar{\varepsilon}\Big]\right\|_{\mathcal{L}(L^2)}\Delta t^3\\
&+\frac{\beta_j T}{48}\|[H_0,\mu]\|_{\mathcal{L}(L^2)}\Delta t^3+\frac{T}{2}\|\mu\|^2_{\mathcal{L}(L^2)}\left(\Delta\varepsilon^2\Delta t+\|\dot{\varepsilon}\|^2_{L^\infty(0,T)}\Delta t^3\right)\\
&+\mathcal{O}(\Delta\varepsilon\Delta t^3)+\mathcal{O}(\Delta t^4).
\end{aligned}$$

The result follows, assuming that $\Delta t,\Delta\varepsilon\leq 1$, with

$$\lambda_1'=T\|\mu\|^2_{\mathcal{L}(L^2)}$$

and

$$\lambda_2'=\frac{\alpha_j T}{24}\left\|\Big[[H_0,\mu],H_0-\mu\bar{\varepsilon}\Big]\right\|_{\mathcal{L}(L^2)}+\frac{\beta_j T}{48}\|[H_0,\mu]\|_{\mathcal{L}(L^2)}+\frac{T}{2}\|\mu\|^2_{\mathcal{L}(L^2)}\|\dot{\varepsilon}\|^2_{L^\infty(0,T)}$$

□

In this theorem, the constants λ_2' depends on $\|\varepsilon\|_{L^\infty(0,T)}$ through the commutator $\Big[[H_0,\mu],H_0-\mu\bar{\varepsilon}\Big]$ that appears in (30). This contrasts with the result obtained in Theorem 1. The explanation of this situation comes from the fact that the norms of $\varphi_j(s):=S_j(t_{j+1}-s)\mu S_j(s-t_j)$ (defined in (18)) and its first derivative does not depend on $\|\varepsilon\|_{L^\infty(0,T)}$, whereas its second derivative does. Thus, errors in Algorithm 2 depend on L^∞-norm of the control field as in the case of the second order Strang operator splitting. Although these two methods present the same computational complexity, the order of Algorithm 2 is higher when $\Delta\varepsilon$ scales Δt^3.

5. Improvement in the limit of large intensities

We now describe a way to improve the time order of the Algorithm 1 in the case of large intensities. The following method enables to replace $\Delta\varepsilon$ by $\Delta\varepsilon\Delta t$ in the estimates.

5.1. **Algorithm.** The algorithm we propose improves the accuracy in the approximation of ε. This improvement is obtained by using two "toolkit" elements instead of one at each time step.

Algorithm 3. *(Improved "toolkit" method for large intensities)*

(1) *Preprocessing. Precompute the "toolkit", i.e. the set of propagators:*

$$S_\ell(\Delta t) \text{ for } \ell=0,\cdots,m,$$

where $(S_\ell(t))_{t\in\mathbb{R}}$ denotes the one-parameter group generated by the operator $H_0-\mu\bar{\varepsilon}_\ell$, the sequence $(\varepsilon_\ell)_{\ell=0,\cdots,m}$, being defined by (5).

(2) *Given a control field $\varepsilon\in L^\infty$ satisfying $\mathcal{H}$ and $\psi_0^{JK}=\psi_0$, the sequence $(\psi_j^{JK})_{j=0,\dots,N}$ that approximates $(\psi(t_j))_{j=0,\dots,N}$, is obtained recursively by iterating the following loop:*

(a) *Find ℓ_j such that:*

$$\varepsilon(t_{j+1/2})\in[\bar{\varepsilon}_{\ell_j},\bar{\varepsilon}_{\ell_j+1}].$$

(b) *Compute* α_j *and* β_j *such that:*

$$\begin{aligned}\alpha_j\bar{\varepsilon}_{\ell_j}+\beta_j\bar{\varepsilon}_{\ell_j+1} &= \varepsilon(t_{j+1/2})\\ \alpha_j+\beta_j &= 1\end{aligned} \tag{31}$$

(c) *Set* $\psi_{j+1}^{JK}=S_{\ell_j+1}(\Delta t)^{\beta_j}S_{\ell_j}(\Delta t)^{\alpha_j}\psi_j^{JK}$.

In this method, one must perform two online matrices exponentiations. The cost of the corresponding step, namely Step *2c* can be reduced to three matrix products when precomputing the mappings between the diagonalization basis of two consecutive "toolkit" elements.

Remark 5. *Another way to reduce the cost of this step, is to quantify the values of* α_j *(and* β_j*) and precompute a "toolkit" containing elements of the form :* $S_{\ell_j+1}(\Delta t)^{\beta_j}S_{\ell_j}(\Delta t)^{\alpha_j}$. *This method is tested in Sec. 6.*

5.2. **Analysis of the method.** We can now repeat the analysis that has been done in the proof of Theorem 1 to obtain the following estimate.

Theorem 3. *Let* $\varepsilon\in W^{3,\infty}(0,T)$, ψ *be the corresponding solution of* (3) *and* ψ^{JK} *the approximation of* ψ *obtained with Algorithm 2. There exists* $\lambda_1''>0$, $\lambda_2''>0$, *both independent of* $\|\varepsilon\|_{L^\infty(0,T)}$ *such that, for every* $\Delta t,\Delta\varepsilon\leq 1$,

$$\|\psi(T)-\psi^{JK}(T)\|_{L^2}\leq\lambda_1''\Delta\varepsilon\Delta t+\lambda_2''\Delta t^2.$$

Proof. In this algorithm, two control fields are involved successively in the propagation over the interval $[t_j,t_{j+1}]$. As in the previous proofs, we introduce $\delta(s)=\varepsilon(s)-\bar{\varepsilon}(s)$, with

$$\bar{\varepsilon}(s)=\begin{cases}\bar{\varepsilon}_{\ell_j} & s\in[t_j,t_j+\alpha_j\Delta t[,\\ \bar{\varepsilon}_{\ell_j+1} & s\in[t_j+\alpha_j\Delta t,t_{j+1}[.\end{cases}$$

Note first that for all $s\in[t_j,t_{j+1}]$

$$|\delta(s)|\leq\Delta\varepsilon+\frac{1}{2}\|\dot{\varepsilon}\|_{L^\infty(0,T)}\Delta t \tag{32}$$

and denote by $(S_j(t))_{j=0,\ldots,N-1}$ and $\left(S_j'(t)\right)_{j=0,\ldots,N-1}$ the one-parameter semi-groups generated by the operators $H_0-\mu\bar{\varepsilon}_{\ell_j}$ and $H_0-\mu\bar{\varepsilon}_{\ell_j+1}$ respectively.

Following the same analysis as for Algorithm 1, we set $(\psi_j^{JK})_{j=0,\ldots,N}$ as the time discretization of the solution of:

$$\begin{cases} i\partial_t\psi^{JK}(t)=\left(H_0-\mu\bar{\varepsilon}(t)\right)\psi^{JK}(t), & \mathbb{R}^\gamma\times(0,T)\\ \psi^{JK}(0)=\psi_0, & \mathbb{R}^\gamma\end{cases} \tag{33}$$

where $\bar{\varepsilon}(t)$ (defined right above) is constant over each interval $[t_j,t_j+\alpha_j\Delta t[$ and $[t_j+\alpha_j\Delta t,t_{j+1}[$, with $j=0,...,N-1$. In the same way as we obtained (9), the solution ψ of (3) satisfies,

$$\psi(t_j+\alpha_j\Delta t)=S_j(\alpha_j\Delta t)\psi(t_j)-i\int_{t_j}^{t_j+\alpha_j\Delta t}S_j(t_j+\alpha_j\Delta t-s)\mu\delta(s)\psi(s)\,ds$$

and

$$\psi(t_{j+1})=S_j'(\beta_j\Delta t)\psi(t_j+\alpha_j\Delta t)-i\int_{t_j+\alpha_j\Delta t}^{t_{j+1}}S_j'(t_{j+1}-s)\mu\delta(s)\psi(s)\,ds.$$

As in (13), it gives rise to:

$$
\begin{aligned}
&\psi(t_{j+1}) - S'_j(\beta_j\Delta t)S_j(\alpha_j\Delta t)\psi(t_j)\\
= \;& i\int_{t_j+\alpha_j\Delta t}^{t_{j+1}} S'_j(t_{j+1}-s)\mu\delta(s)\psi(s)\; ds\\
&+i\int_{t_j}^{t_j+\alpha_j\Delta t} S'_j(\beta_j\Delta t)S_j(t_j+\alpha_j\Delta t-s)\mu\delta(s)\psi(s)\; ds\\
= \;& i\int_{t_j+\alpha_j\Delta t}^{t_{j+1}} S'_j(t_{j+1}-s)\mu\delta(s)\left(\psi(s)-S'_j(s-t_j-\alpha_j\Delta t)\psi(t_j+\alpha_j\Delta t)\right)\; ds\\
&+i\int_{t_j}^{t_j+\alpha_j\Delta t} S'_j(\beta_j\Delta t)S_j(t_j+\alpha_j\Delta t-s)\mu\delta(s)\left(\psi(s)-S_j(s-t_j)\psi(t_j)\right)\; ds\\
&+i\int_{t_j+\alpha_j\Delta t}^{t_{j+1}} \delta(s)\widetilde{\varphi}'_j(s)S_j(\alpha_j\Delta t)\psi(t_j)\; ds\\
&+i\int_{t_j}^{t_j+\alpha_j\Delta t} S'_j(\beta_j\Delta t)\delta(s)\widetilde{\varphi}_j(s)\psi(t_j)\; ds.
\end{aligned}
\tag{34}
$$

where $\widetilde{\varphi}'_j(s) := S'_j(t_{j+1}-s)\mu S'_j(s-t_j-\alpha_j\Delta t)$ and $\widetilde{\varphi}_j(s) := S_j(t_j+\alpha_j\Delta t-s)\mu S_j(s-t_j)$. As in the proof of Theorem 1 (see right above (12)) we use the appropriate decomposition

$$
\begin{aligned}
&\psi(T)-\psi^{JK}(T) = \psi(T) - S'_{N-1}(\beta_{N-1}\Delta t)S_{N-1}(\alpha_{N-1}\Delta t)\psi(t_{N-1})\\
&+\sum_{j=0}^{N-2} S'_{N-1}(\beta_{N-1}\Delta t)S_{N-1}(\alpha_{N-1}\Delta t)\dots S'_{j+1}(\beta_{j+1}\Delta t)S_{j+1}(\alpha_{j+1}\Delta t)\\
&\qquad\qquad\times\left(\psi(t_{j+1}) - S'_j(\beta_j\Delta t)S_j(\alpha_j\Delta t)\psi(t_j)\right)\\
&+S'_{N-1}(\beta_{N-1}\Delta t)S_{N-1}(\alpha_{N-1}\Delta t)\dots S'_0(\beta_0\Delta t)S_0(\alpha_0\Delta t)\psi_0 - \psi^{JK}(T)
\end{aligned}
$$

where the last line is equal to 0 since ψ^{JK} satisfies (33) on $[0,T]$. We have the corresponding estimate (see (12))

$$
\|\psi(T)-\psi^{JK}(T)\|_{L^2} \le \sum_{j=0}^{N-1}\left\|\psi(t_{j+1}) - S'_j(\beta_j\Delta t)S_j(\alpha_j\Delta t)\psi(t_j)\right\|_{L^2}
$$

and we will thus calculate and estimate in L^2-norm for all j the four terms of (34). As in (16), but using now the new estimate (32) of δ, the two first terms of the right hand side of (34) can be respectively estimated by:

$$
\begin{aligned}
&\left\| i\int_{t_j+\alpha_j\Delta t}^{t_{j+1}} S'_j(t_{j+1}-s)\mu\delta(s)\left(\psi(s)-S'_j(s-t_j-\alpha_j\Delta t)\psi(t_j+\alpha_j\Delta t)\right)\; ds\right\|_{L^2}\\
&\qquad\le\; \beta_j\|\mu\|^2_{\mathcal{L}(L^2)}\left(\Delta\varepsilon^2\Delta t^2 + \frac{1}{2}\|\dot\varepsilon\|^2_{L^\infty(0,T)}\Delta t^4\right)
\end{aligned}
\tag{35}
$$

and

$$
\begin{aligned}
&\left\| i\int_{t_j}^{t_j+\alpha_j\Delta t} S'_j(\beta_j\Delta t)S_j(t_j+\alpha_j\Delta t-s)\mu\delta(s)\left(\psi(s)-S_j(s-t_j)\psi(t_j)\right)\right\|_{L^2}\\
&\qquad\le\; \alpha_j\|\mu\|^2_{\mathcal{L}(L^2)}\left(\Delta\varepsilon^2\Delta t^2 + \frac{1}{2}\|\dot\varepsilon\|^2_{L^\infty(0,T)}\Delta t^4\right).
\end{aligned}
\tag{36}
$$

Let us now focus on the third and fourth terms of (34). We have:

$$\begin{aligned}\int_{t_j+\alpha_j\Delta t}^{t_{j+1}} \delta(s)\widetilde{\varphi}_j'(s)S_j(\alpha_j\Delta t)\psi(t_j)\ ds = &\int_{t_j+\alpha_j\Delta t}^{t_{j+1}} \delta(s)\widetilde{\varphi}_j'(t_j+\alpha_j\Delta t)S_j(\alpha_j\Delta t)\psi(t_j)\ ds\\ &+\int_{t_j+\alpha_j\Delta t}^{t_{j+1}} \delta(s)\int_{t_j+\alpha_j\Delta t}^{s}\partial_u\widetilde{\varphi}_j'(u)\ du\ S_j(\alpha_j\Delta t)\psi(t_j)\ ds\\ =&\int_{t_j+\alpha_j\Delta t}^{t_{j+1}} \delta(s)S_j'(\beta_j\Delta t)\mu S_j(\alpha_j\Delta t)\psi(t_j)\ ds\\ &+\int_{t_j+\alpha_j\Delta t}^{t_{j+1}} \delta(s)\int_{t_j+\alpha_j\Delta t}^{s}\partial_u\widetilde{\varphi}_j'(u)\ du\ S_j(\alpha_j\Delta t)\psi(t_j)\ ds\end{aligned}$$

and

$$\begin{aligned}\int_{t_j}^{t_j+\alpha_j\Delta t} S_j'(\beta_j\Delta t)\delta(s)\widetilde{\varphi}_j(s)\psi(t_j)\ ds = &\int_{t_j}^{t_j+\alpha_j\Delta t}\delta(s)S_j'(\beta_j\Delta t)\widetilde{\varphi}_j(t_j+\alpha_j\Delta t)\psi(t_j)\ ds\\ &-\int_{t_j}^{t_j+\alpha_j\Delta t}\delta(s)S_j'(\beta_j\Delta t)\int_s^{t_j+\alpha_j\Delta t}\partial_u\widetilde{\varphi}_j(u)\ du\ \psi(t_j)\ ds\\ =&\int_{t_j}^{t_j+\alpha_j\Delta t}\delta(s)S_j'(\beta_j\Delta t)\mu S_j(\alpha_j\Delta t)\psi(t_j)\ ds\\ &-\int_{t_j}^{t_j+\alpha_j\Delta t}\delta(s)S_j'(\beta_j\Delta t)\int_s^{t_j+\alpha_j\Delta t}\partial_u\widetilde{\varphi}_j(u)\ du\ \psi(t_j)\ ds.\end{aligned}$$

By means of (31), we have:

$$\begin{aligned}\int_{t_j}^{t_{j+1}}\delta(s)\ ds &= \int_{t_j}^{t_{j+1}}\varepsilon(s)-\varepsilon(t_{j+1/2})\ ds+\int_{t_j}^{t_{j+1}}\varepsilon(t_{j+1/2})-\bar{\varepsilon}(s)\ ds\\ &= \int_{t_j}^{t_{j+1}}\varepsilon(s)-\varepsilon(t_{j+1/2})\ ds\\ &= \int_{t_j}^{t_{j+1}}\ddot{\varepsilon}(\theta(s))\frac{1}{2}(s-t_{j+1/2})^2\ ds,\end{aligned}$$

where $\theta(s)\in[t_j,t_{j+1}]$. Consequently,

$$\left\|\int_{t_j}^{t_{j+1}}\delta(s)S_j'(\beta_j\Delta t)\mu S_j(\alpha_j\Delta t)\psi(t_j)\ ds\right\|_{L^2}\leq\frac{1}{24}\|\mu\|_{\mathcal{L}(L^2)}\|\ddot{\varepsilon}\|_{L^\infty(0,T)}\Delta t^3. \tag{37}$$

From (21) and (32), we obtain

$$\begin{aligned}&\left\|\int_{t_j+\alpha_j\Delta t}^{t_{j+1}}\delta(s)\int_{t_j+\alpha_j\Delta t}^{s}\partial_u\widetilde{\varphi}_j'(u)\ du\ S_j(\alpha_j\Delta t)\psi(t_j)\ ds\right\|_{L^2}\\ &\qquad\leq\frac{1}{2}\beta_j^2\|[H_0,\mu]\|_{\mathcal{L}(L^2)}\left(\Delta\varepsilon+\frac{1}{2}\|\dot{\varepsilon}\|_{L^\infty(0,T)}\Delta t\right)\Delta t^2\end{aligned} \tag{38}$$

and similarly, we find that:

$$\left\| \int_{t_j}^{t_j+\alpha_j\Delta t} \delta(s) S_j'(\beta_j\Delta t) \int_s^{t_j+\alpha_j\Delta t} \partial_u \widetilde{\varphi}_j(u)\ du\ \psi(t_j)\ ds \right\|_{L^2} \leq \frac{1}{2}\alpha_j^2 \|[H_0,\mu]\|_{\mathcal{L}(L^2)} \left(\Delta\varepsilon + \frac{1}{2}\|\dot\varepsilon\|_{L^\infty(0,T)}\Delta t \right) \Delta t^2. \tag{39}$$

Combining (35), (36), (37), (38) and (39), we obtain:

$$\begin{aligned} \left\|\psi(t_{j+1}) - S_j'(\beta_j\Delta t) S_j(\alpha_j\Delta t)\psi(t_j)\right\|_{L^2} \leq\ & \|\mu\|^2_{\mathcal{L}(L^2)} \left(\Delta\varepsilon^2 + \frac{1}{2}\|\dot\varepsilon\|^2_{L^\infty(0,T)}\Delta t^2 \right) \Delta t^2 \\ & + \frac{1}{24}\|\mu\|_{\mathcal{L}(L^2)} \|\ddot\varepsilon\|_{L^\infty(0,T)}\Delta t^3 \\ & + \frac{1}{2}\|[H_0,\mu]\|_{\mathcal{L}(L^2)} \left(\Delta\varepsilon + \frac{1}{2}\|\dot\varepsilon\|_{L^\infty(0,T)}\Delta t \right) \Delta t^2. \end{aligned}$$

The global estimate follows

$$\begin{aligned} \|\psi(T) - \psi^{JK}(T)\|_{L^2} \leq\ & \sum_{j=0}^{N-1} \left\|\psi(t_{j+1}) - S_j'(\beta_j\Delta t) S_j(\alpha_j\Delta t)\psi(t_j)\right\|_{L^2} \\ \leq\ & \|\mu\|^2_{\mathcal{L}(L^2)} \left(\Delta\varepsilon^2 + \frac{1}{2}\|\dot\varepsilon\|^2_{L^\infty(0,T)}\Delta t^2 \right) T\Delta t \\ & + \frac{1}{24}\|\mu\|_{\mathcal{L}(L^2)} \|\ddot\varepsilon\|_{L^\infty(0,T)} T\Delta t^2 \\ & + \frac{1}{2}\|[H_0,\mu]\|_{\mathcal{L}(L^2)} \left(\Delta\varepsilon + \frac{1}{2}\|\dot\varepsilon\|_{L^\infty(0,T)}\Delta t \right) T\Delta t \end{aligned}$$

and the proof of Theorem 3 is complete, assuming that $\Delta t, \Delta\varepsilon \leq 1$, with

$$\lambda_1'' = \frac{1}{2}\|[H_0,\mu]\|_{\mathcal{L}(L^2)}T + \|\mu\|^2_{\mathcal{L}(L^2)}T,$$

$$\begin{aligned} \lambda_2'' =\ & \frac{1}{4}\|[H_0,\mu]\|_{\mathcal{L}(L^2)}\|\dot\varepsilon\|_{L^\infty(0,T)}T + \frac{1}{24}\|\mu\|_{\mathcal{L}(L^2)}\|\ddot\varepsilon\|_{L^\infty(0,T)}T \\ & + \frac{1}{2}\|\mu\|^2_{\mathcal{L}(L^2)}\|\dot\varepsilon\|_{L^\infty(0,T)}T. \end{aligned}$$

□

6. Numerical results

In this section, we check numerically that the order of the estimates we have obtained in this paper are optimal, and we compare computational costs of the methods.

6.1. **Model.** In order to test the performance of the algorithms on a realistic case, a model already treated in the literature has been considered. The system is a molecule of HCN modeled as a rigid rotator. We refer the reader to [20, 36] for numerical details concerning this system. As a control field, we use an arbitrary field of the form $\varepsilon(t) = \varepsilon_{\max}\sin(\omega t)$, with $\varepsilon_{\max} = 5.10^{-5}$ and $\omega = 5.10^{-6}$. The parameters are chosen in accordance with usual scales considered for this model. The use of an analytic formula for the field enables us to work with exact values, i.e. to test the cases $\Delta\varepsilon = 0$.

6.2. **Orders of convergence.** To test the time order, we first work with $\Delta\varepsilon = 0$, with various values of Δt. The numerical orders correspond to the ones obtained in our analysis. Curves of convergence are depicted in Fig. 1.

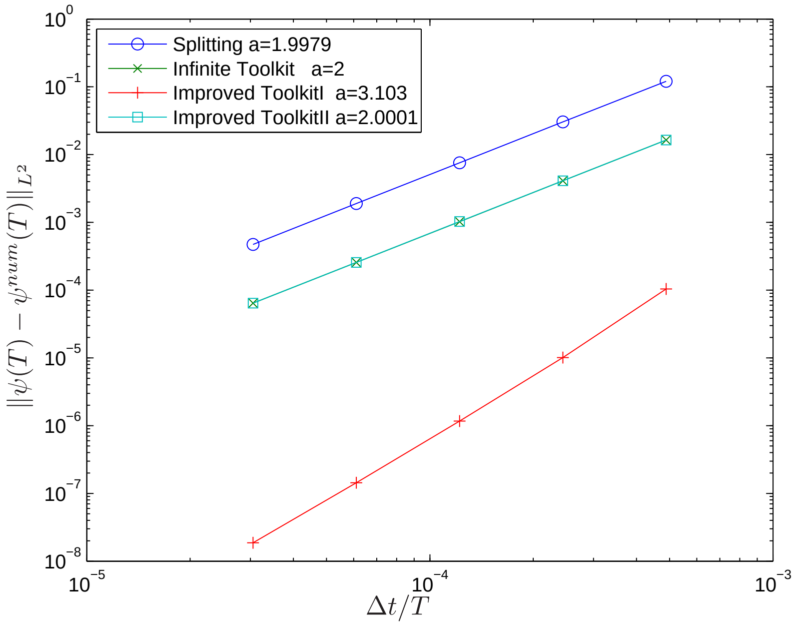

FIGURE 1. Error with respect to Δt, when $\Delta\varepsilon = 0$ for "toolkit" method and Improved "toolkit" I method, and when$\Delta\varepsilon = c\Delta t$ for Improved "toolkit" II method. Here, ψ^{num} stands for the approximation of ψ when using the "toolkit" method, the second order Strang operator splitting, the Improved "toolkit" I method and the Improved "toolkit" II. The coefficient a is the regression coefficient.

The order with respect to $\Delta\varepsilon$ is also obtained numerically by using a small time step. In this test, the numerical order is consistent with the one obtained in Theorem 1. The convergence with respect to this parameter is presented in Fig. 2.

6.3. **Computational cost.** In a second test, we compare the computational costs of the methods. To do this, we look for the values of $N = \frac{T}{\Delta t}$ and $m = \frac{\varepsilon_{\max}}{\Delta\varepsilon}$ that enable to reach a fixed arbitrary error of $Tol = 5.10^{-3}$ (recall that in any case the error cannot exceed 2). For sake of simplicity, we only test powers of 2. In this test, we also include the quantified version of the Improved "toolkit" II which is described in Remark 5. In our case, the parameters α and β were quantified among 100 values uniformly distributed in $[0, 1]$.

These tests show that "toolkit" methods always give better results as the second order Strang operator splitting.

The two improvements we propose in this paper enable to reduce respectively the global number of matrix products and the size of the "toolkit", which is in agreement with the analysis we have done. Note that the second improvement reduce significantly preprocessing step. This fact makes feasible the quantified version of it, which requires intrinsically a larger "toolkit".

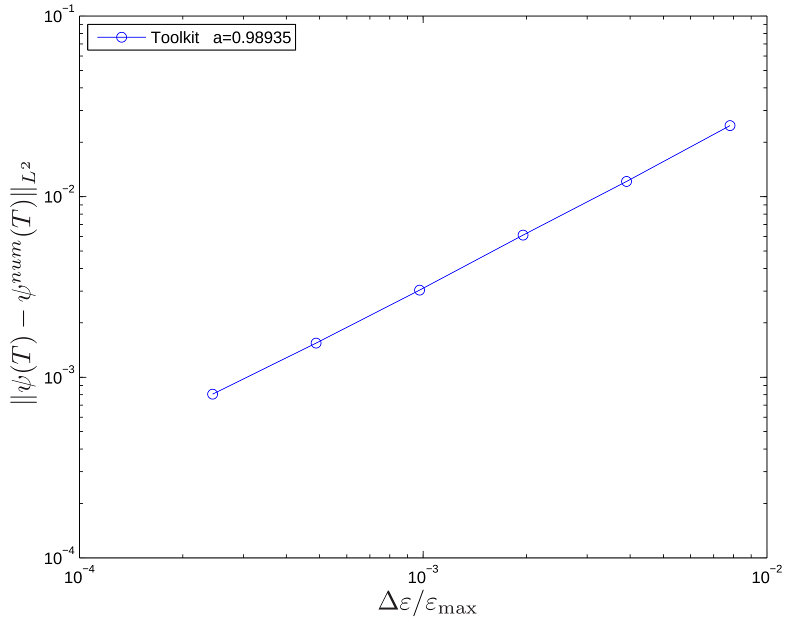

FIGURE 2. Error with respect to $\Delta\varepsilon$, when Δt is small. Here, ψ^{num} stands for the approximation of ψ when using the "toolkit" method.

	$N = \frac{T}{\Delta t}$	Matrix products	$m = \frac{\varepsilon_{\max}}{\Delta\varepsilon}$
Strang Op. Splitting	16384	32768	-
Toolkit	8192	8192	16384
Improved "toolkit" I	1024	2048	16384
Improved "toolkit" II	4096	12288	16
Quantified Improved "toolkit" II	4096	4096	6400

TABLE 1. Values of numerical parameters corresponding to a tolerance error of $Tol = 5.10^{-3}$.

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