Sixth-order factorization of the evolution operator for time-dependent potentials

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The evolution operator of a quantum system in a time-dependent potential is factorized in unitary exponential operators at order 6. This expression is derived with the time-ordering method. It is compared with lower-order factorizations on several simple one-dimensional examples. Better accuracies are reached at sixth order for a given time step than at lower orders. Due to a significant increase of computation duration per time step, the sixth-order approximation is mainly useful when high accuracies are required.

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I. INTRODUCTION

Solving the time-dependent Schrödinger equation is important in many subfields of quantum physics. Our group is involved in numerical resolutions of a three-dimensional time-dependent Schrödinger equation for studying the breakup of one-neutron halo nuclei [1]. A halo nucleus is interpreted as a normal nucleus surrounded by one or two distant neutrons [2]. Future breakup studies will crucially depend on improvements of the efficiency of algorithms involving time-dependent potentials. We think that such improvements will also be useful for other atomic or nuclear problems involving a time-dependent Schrödinger equation with three spatial dimensions.

The time-dependent Hamiltonian is the sum of a time-independent part H_0, containing the kinetic energy and a possible time-independent part of the potential energy, and a time-dependent potential $V(t)$. The basic idea of many algorithms is to apply an approximation of the evolution operator depending on a time step Δt to a wave function described in a representation where the time-dependent potential matrix is diagonal. This is the case in finite-difference methods [3], but also in the discrete-variable representation [4] or its accurate subset the Lagrange-mesh method [5]. The evolution operator is approximated as a product of unitary exponential operators depending either on H_0 or on V evaluated at some time. The accuracy of this approximation is expressed as a given order k corresponding to an error of order Δt^{k+1} per time step. For symmetric factorizations, order k is even [6,7].

Most calculations are based on variants of a second-order factorization [see Eq. (2) below]. The approximate evolution operator contains one exponential factor involving H_0, whose application on the wave function represents the major part of the computation time. A number of higher-order approximations have been proposed. The first fourth-order approximation has been derived by Bandrauk and Shen [8]. Suzuki has introduced a general symbolic scheme to deal with the time-ordered exponential expression of the evolution operator [9]. Chin and Chen [10], starting from the symbolic representation of the evolution operator proposed by Suzuki, derived several variants of fourth-order factorizations. In Ref. [11], it was shown that fourth-order approximations can also be obtained from the Magnus expansion of the evolution operator [12] complemented by consistent quadrature approximations of integrals. Other factorization schemes exist, which do not separate H_0 and $V(t)$ [13].

The aim of the present work is to derive a sixth-order algorithm and to evaluate its interest with respect to the simpler second- and fourth-order algorithms. The approach based on the Magnus expansion could only be extended to sixth order for simple form factors of potential $V(t)$. Therefore, we turn to the technique proposed by Suzuki [9] and make use of recent results on the factorization of exponential operators [14]. Sixth-order factorizations, among others, are derived, classified, and discussed in Ref. [15]. The authors also evaluate the relative efficiencies of time-dependent algorithms on classical problems. Here, the comparative merits of different algorithms are evaluated on simple time-dependent quantum test cases: the harmonic oscillator forced by an electric pulse or an oscillating electric field, which are exactly solvable [16,17], and the Walker-Preston model [18], which is well documented [10].

In Sec. II, we recall different unitary approximations of the evolution operator where H_0 and $V(t)$ are separated. A sixth-order algorithm is presented in Sec. III, and tested on simple examples in Sec. IV. Concluding remarks are presented in Sec. V.

II. APPROXIMATIONS UP TO ORDER 4

When the potential depends on time, the Hamiltonian can be written as

$$H(t) = H_0 + V(t), \tag{1}$$

where H_0 may contain a time-independent potential term V_0 in addition to the kinetic energy T. In the following, we only assume that $V(t)$ commutes with itself at different times, and with V_0. For a given time step Δt, we are interested in unitary factorizations of the evolution operator $U(t+\Delta t, t)$, generally denoted as U in the following, where H_0 and $V(t)$ are separated.

Many practical calculations are based on the second-order factorization,

$$U = e^{-i(1/2)\Delta t V(t+\Delta t-\alpha\Delta t)} e^{-i\Delta t H_0} e^{-i(1/2)\Delta t V(t+\alpha\Delta t)} + O(\Delta t^3), \tag{2}$$

with $\alpha \in [0,1]$. The most interesting cases are $\alpha = 1/2$, which leads to a symmetric expression, and $\alpha = 0$, which allows grouping the first exponential operator with the last factor of the next iteration.

Following the technique proposed by Suzuki in Ref. [9], which is described in the next section, Chin and Chen [10] have derived the fourth-order approximation,

$$U = e^{-i(1/6)\Delta t V(t+\Delta t)} e^{-i(1/2)\Delta t H_0} e^{-i(2/3)\Delta t \tilde{V}_0(t+(1/2)\Delta t)} \times e^{-i(1/2)\Delta t H_0} e^{-i(1/6)\Delta t V(t)} + O(\Delta t^5), \tag{3}$$

with the gradient term [19,14]

$$\tilde{V}_i(t) = V(t) - \frac{d_i}{m}\Delta t^2 [\nabla V(t)]^2, \tag{4}$$

where m is the mass of the particle and $d_0 = 1/48$.

Omelyan, Mryglod, and Folk have shown that smaller errors can be expected at fourth order at the cost of additional exponential factors [15]. Also with Suzuki's technique, they propose in Eqs. (48) and (53) the fourth-order approximation,

$$\begin{aligned} U = {} & e^{-iv_0\Delta t \tilde{V}_0(t+\Delta t)} e^{-i\Delta t_0 H_0} e^{-iv_1\Delta t V(t+\Delta t-\Delta t_0)} \\ & \times e^{-i\Delta t_1 H_0} e^{-iv_2\Delta t \tilde{V}_2[t+(1/2)\Delta t]} e^{-i\Delta t_1 H_0} e^{-iv_1\Delta t V(t+\Delta t_0)} \\ & \times e^{-i\Delta t_0 H_0} e^{-iv_0\Delta t \tilde{V}_0(t)} + O(\Delta t^5). \end{aligned} \tag{5}$$

The coefficients of this decomposition are

$$\begin{aligned} \Delta t_0 &= 0.192\,112\,527\,742\,946\,4\ \Delta t, \quad \Delta t_1 = \frac{1}{2}\Delta t - \Delta t_0, \\ v_0 &= 0.058\,518\,726\,134\,556\,21, \\ v_1 &= 0.285\,216\,224\,068\,709\,1, \quad v_2 = 1 - 2(v_0 + v_1), \\ d_0 &= 0.007\,415\,743\,802\,826\,3, \\ d_2 &= 0.007\,767\,172\,706\,950\,9. \end{aligned} \tag{6}$$

It is interesting to see whether the increase of computing time is compensated by the expected improvement.

In Ref. [11], we have shown that it is possible to derive approximations at order 4 from the Magnus expansion [12]. As a first step, a factorization is obtained where H_0 and V are separated,

$$U = e^{-i(1/6)\Delta t W_1 + i\Delta t^2 W_2} e^{-i(1/2)\Delta t H_0} e^{-i(2/3)\Delta t \tilde{W}_1} \times e^{-i(1/2)\Delta t H_0} e^{-i(1/6)\Delta t W_1 - i\Delta t^2 W_2} + O(\Delta t^5). \tag{7}$$

It involves integrals of the potential

$$W_i(t) = \frac{1}{\Delta t^{2i-1}} \int_t^{t+\Delta t} dt' \left(t + \frac{1}{2}\Delta t - t' \right)^{i-1} V(t') dt' \tag{8}$$

and a modified integral $\tilde{W}_1$ defined as in Eq. (4). Then, these integrals can be approximated in a way consistent with the expected accuracy. In Ref. [11], we proposed to approximate the integrals W_1 and W_2 with the Simpson formula [3] as

$$W_1(t) = \frac{1}{6}\left[V(t) + 4V\left(t + \frac{1}{2}\Delta t \right) + V(t+\Delta t) \right] + O(\Delta t^4) \tag{9}$$

and

$$W_2(t) = \frac{1}{12\Delta t}[V(t) - V(t+\Delta t)] + O(\Delta t^2). \tag{10}$$

As in Eq. (3), it only requires three evaluations of the potential. This number can be reduced to 2 with a two-point Gauss quadrature formula [3],

$$W_1(t) = \frac{1}{2}\left[V\left(t + \frac{3-\sqrt{3}}{6}\Delta t \right) + V\left(t + \frac{3+\sqrt{3}}{6}\Delta t \right) \right] + O(\Delta t^4) \tag{11}$$

and

$$W_2(t) = \frac{\sqrt{3}}{12\Delta t}\left[V\left(t + \frac{3-\sqrt{3}}{6}\Delta t \right) - V\left(t + \frac{3+\sqrt{3}}{6}\Delta t \right) \right] + O(\Delta t^2). \tag{12}$$

This gain is, however, only apparent when the process is iterated, since the first exponential in Eq. (7) can be grouped with the last exponential of the next iteration. However, this variant appears to be slightly more precise in the examples of Sec. IV.

III. AN APPROXIMATION AT ORDER 6

We have tried to generalize the Magnus approach up to order 6, but we could only achieve it in particular cases. Some commutators appearing in integrals of the Magnus expansion, such as $\mathbf{[}[H_0, V(t')], [H_0, V(t'')]\mathbf{]}$ could not be disentangled in factorized exponentials depending either on H_0 or on V. These terms vanish in the examples considered below, but do not in more general cases. So, we turned to the Suzuki approach using time-ordered exponentials [9] applied by Chin and Chen at order 4 [10]. It allows transforming a decomposition at a given order for time-independent potentials in a decomposition valid for time-dependent potentials.

Following Ref. [9], the evolution operator can be written for time-dependent potentials as

$$U(t+\Delta t, t) = e^{-i\Delta t[H(t)+iD]} = e^{-i\Delta t[(H_0+iD)+V(t)]}, \tag{13}$$

where D is the forward time derivative operator

$$D = \frac{\overleftarrow{\partial}}{\partial t}. \tag{14}$$

This operator possesses the property,

$$F(t) e^{\Delta t D} G(t) = F(t+\Delta t) G(t), \tag{15}$$

i.e., it translates in time the operators located at its left.

Property (15) allows deriving an approximation of the evolution operator in the following way. A decomposition of

$\exp[-i\Delta t(A+B)]$ with $A=H_0+iD$ and $B=V(t)$ is chosen at some order and applied to Eq. (13). The commutation property $[H_0,D]=0$ implies

$$e^{-i\Delta t(H_0+iD)} = e^{-i\Delta t H_0} e^{\Delta t D}. \tag{16}$$

Double commutators of type $[V(t),[H_0+iD,V(t)]]$ appear beyond second order. These commutators are simplified by using the commutation property [10]

$$[V(t),[D,V(t)]] = 0 \tag{17}$$

and the gradient expression [19,14]

$$[V(t),[H_0,V(t)]] = \frac{1}{m}[\nabla V(t)]^2. \tag{18}$$

Then, relation (15) is employed. With a fourth-order decomposition, this procedure leads to Eq. (3) [10].

Let us apply this procedure to the optimal decomposition at sixth order of $\exp[-i\Delta t(A+B)]$ presented in Eq. (50) of Ref. [14]. It is optimal in the sense that the authors of Ref. [14] have selected it among various possibilities to minimize the error. The above procedure leads to

$$\begin{aligned} U(t+\Delta t,t) &= e^{-i\Delta t_0 H_0} e^{-iv_0\Delta t \tilde{V}_0(t+\Delta t-\Delta t_0)} e^{-i\Delta t_1 H_0} e^{-iv_1\Delta t V(t+\Delta t-\Delta t_1-\Delta t_0)} \\ &\times e^{-i\Delta t_2 H_0} e^{-iv_2\Delta t \tilde{V}_2[t+(1/2)\Delta t]} e^{-i\Delta t_2 H_0} e^{-iv_1\Delta t V(t+\Delta t_1+\Delta t_0)} \\ &\times e^{-i\Delta t_1 H_0} e^{-iv_0\Delta t \tilde{V}_0(t+\Delta t_0)} e^{-i\Delta t_0 H_0} + O(\Delta t^7), \end{aligned} \tag{19}$$

with $\tilde{V}_i$ defined in Eq. (4). The coefficients of this decomposition are

$$\begin{aligned} \Delta t_0 &= 0.109\,705\,972\,394\,868\,2\ \Delta t, \\ \Delta t_1 &= 0.414\,063\,226\,731\,083\,1\ \Delta t, \\ \Delta t_2 &= \frac{1}{2}\Delta t - \Delta t_1 - \Delta t_0, \\ v_0 &= 0.269\,331\,584\,893\,530\,1, \\ v_1 &= 1.131\,980\,348\,651\,556, \quad v_2 = 1-2(v_0+v_1), \\ d_0 &= 0.003\,208\,744\,099\,999\,45, \\ d_2 &= 0.007\,348\,391\,794\,869\,08. \end{aligned} \tag{20}$$

Notice that, in quantum-mechanical applications, all coefficients Δt_i or v_i in exponential operators of the chosen decomposition need not be positive to ensure a good accuracy.

Decomposition (19) involves six exponentials of H_0 and is, in this respect, not the most economical at sixth order in Ref. [14]. Indeed factorization (45) of that reference leads to an approximation of the evolution operator containing only four exponentials of H_0. However, we have observed that the corresponding algorithm provides results that are less accurate by about two orders of magnitude. Moreover, it involves evaluations of the potential outside the interval $[t,t+\Delta t]$. This may raise numerical problems in the first and last time intervals when there is no obvious way of extrapolating the potential before the initial time or after the final time. That decomposition is therefore not competitive with decomposition (19) and will not be further considered here. In the examples below, we however discuss another decomposition at order 6, involving more exponentials but without gradient terms.

The first and last exponentials in Eq. (19) can be grouped in the iteration so that the average number of exponentials of H_0 per time step is only five. This number should be compared with the two exponentials in Eqs. (3) and (7) or four exponentials in Eq. (5) at fourth order and with the single exponential at second order. We associate with these decompositions an integer n taking the values 5, 2, 4, and 1, respectively.

IV. APPLICATION TO SIMPLE ONE-DIMENSIONAL SYSTEMS

In order to appreciate the efficiency of the sixth-order algorithm, we compare the different approximations (2), (3), (5), (7), and (19) on two one-dimensional quantum problems. The forced harmonic oscillator can be solved analytically [16,17]. The Walker-Preston model [18] represents a diatomic molecule in a time-dependent electric field and has been studied in Ref. [10]. In a first step, we only compare the accuracies of the algorithms. We check that a sixth-order accuracy is really obtained and discuss the improvement with respect to lower orders. In a second step, we address the utility of the algorithms by comparing their efficiencies, i.e., by taking account the fact that the computer time involved in a single time step differs at the various orders.

The one-dimensional time-dependent Hamiltonian reads

$$H = -\frac{1}{2m}\frac{\partial^2}{\partial x^2} + V_0(x) - f(t)x. \tag{21}$$

The first two terms are interpreted as operator H_0. The last term is the time-dependent potential $V(t)$. A different separation is also briefly discussed below. The wave function $\psi(x,t)$ is represented on a mesh in the spirit of the Lagrange-mesh [5] or discrete-variable [4] methods. Following the discussion in Ref. [11], we select a sinc mesh [20,21], which is based on equally spaced mesh points $x_j=x_1+(j-1)h$, where x_1 and h are scaling parameters and j varies from 1 to N. All potential terms are represented by diagonal matrices evaluated at mesh points. The Hamiltonian operator H_0 is represented by matrix elements

$$H_{0ij} = \begin{cases} m^{-1}(-1)^{i-j}(x_i-x_j)^{-2} & (i\neq j), \\ m^{-1}h^{-2}\pi^2/6 + V_0(x_i) & (i=j), \end{cases} \tag{22}$$

where i and j vary from 1 to N. The time-dependent potential is represented at the Gauss approximation related with sinc functions [21] as

$$V_{ij}(t) = -x_i f(t)\delta_{ij}. \tag{23}$$

The mesh covers an interval $[x_1,x_N]$ outside of which wave functions are negligible. The number N of mesh points is

taken large enough so that the results are stable if N is increased. The exponentials of the diagonal potential matrices are very easy to calculate in all cases. The exponentials of the full H_0 matrices are obtained through a diagonalization which must be performed only once, before the iteration starts. In contrast with the second- and fourth-order algorithms (3) and (7), three such matrices must be stored in the sixth-order algorithm. Another approach, using Padé approximations, has been tested in Ref. [11]. It led to a significant loss of accuracy. At sixth order, a (3,3) Padé approximation consistent with the accuracy of the algorithm should be used. Since the kinetic-energy matrix is full, it would not reduce the computing times.

For the forced oscillator, H_0 is the harmonic-oscillator Hamiltonian with $m=1$ and $V_0(x)=(1/2)x^2$. Its eigenfunctions are denoted as $\varphi_n(x)$ and the corresponding energies as E_n. The general solution of the time-dependent Schrödinger equation for the initial wave packet $\psi(x,0)=\varphi_n(x)$ at $t=0$ is given by [16,17]

$$\psi(x,t) = \varphi_n[x - x_0(t)]\exp[ip_0(t)x]\exp \times\left\{-i\int_0^t [E_n + \mathcal{L}_0(t')]dt'\right\}, \tag{24}$$

where $x_0(t)$ and $p_0(t)$ are the classical coordinate and momentum corresponding to H with initial conditions $x_0(0)=0$ and $p_0(0)=0$, respectively. The Lagrangian-like expression $\mathcal{L}_0=(1/2)\dot{x}_0^2-(1/2)x_0^2$ appears in the phase factor. For the time-dependent potential, we consider two cases which allow a fully analytical treatment. As in Ref. [11], we consider a trigonometric pulse. However, for simplicity, we now define it as

$$f_1(t) = \sin^2\frac{1}{2}\pi t, \quad 0 < t < 2, \tag{25}$$

i.e., we eliminate the two periods with $V(t)=0$ before and after the pulse considered in Ref. [11]. A second time-dependent term is the oscillating field

$$f_2(t) = \sin\ \omega t, \tag{26}$$

with $\omega=0.75$. The corresponding explicit expressions of $x_0(t)$ and $\int_0^t \mathcal{L}_0(t')dt'$ are given in the Appendix. In both cases, the system is at $t=0$ in the ground state of the harmonic oscillator.

For the pulse, the interval is $[-8,+8]$ with $N=50$. The pulse increases until $t=1$ and then decreases until $t=2$, where we analyze the accuracy of the final wave function by comparison with the exact result. The final error with respect to the exact calculation is given by the quadratic-norm distance,

$$\varepsilon = \|\psi^{\text{approx.}}(t) - \psi^{\text{exact}}(t)\| \tag{27}$$

at the end $t=2$ of the pulse (25). In Fig. 1, the final error ϵ is plotted as a function of the time step Δt for different factorizations of the evolution operator.

We can distinguish three different behaviors, as expected. At the second-order approximation (2) with $\alpha=1/2$ (open triangles), the error displays a slope typical of Δt^2 since the number of time steps is proportional to $1/\Delta t$. In Fig. 2 of Ref. [11], the slope corresponds to Δt^3, because the average error per time step is represented. At fourth order, the errors of the different approximations display a slope Δt^4. Approximation (7) with the Simpson quadratures (9) and (10) is less accurate than Chin and Chen's algorithm (3) or factorization (7) with the Gauss quadratures (11) and (12), which are almost equivalent. Algorithm (5) is, however, much more accurate. At sixth order (crosses), the error clearly decreases as Δt^6 down to the computer accuracy. It is remarkable that with only two steps ($\Delta t=1$), the accuracy is better than 0.1% with the sixth-order approximation. We also made the calculation at sixth order with H_0 equal to the kinetic energy T while $V(t)$ is $(1/2)x^2-f(t)x$. This case is more general because the gradient terms (4) still depend on x. The results would display no visible difference with respect to Fig. 1. We also test the best gradient-free algorithm (82) and (83) of Ref. [15]. The obtained results (pluses) are slightly less good.

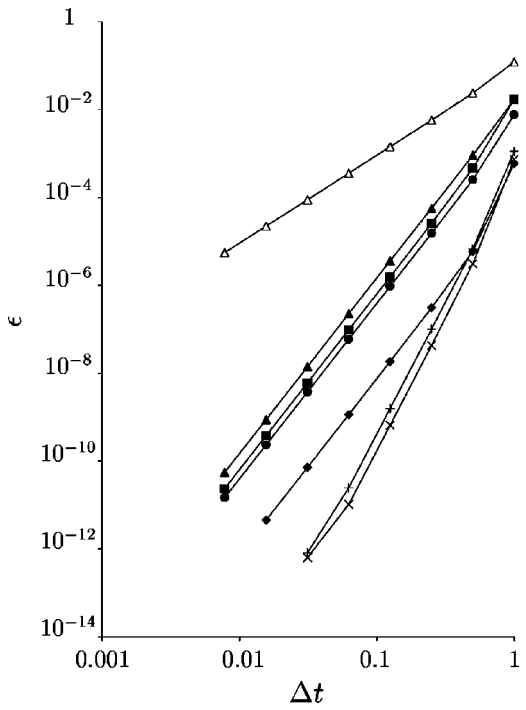

FIG. 1. Accuracies ϵ in different approximations of the time evolution operator as a function of the time step Δt for the harmonic oscillator forced by pulse (25). Second-order factorization (2), open triangles. Fourth-order factorizations (7) with Simpson integration, full triangles; (7) with Gauss integration, full circles; (3) full squares; and (5) full diamonds. Sixth-order factorization (19), crosses, and gradient-free factorization (see text), pluses.

For the oscillating field, we choose the spatial interval $[-12,+12]$ with $N=75$. The natural time unit is the oscillation period $\tau=2\pi/\omega$. In this case, we calculate the error (27) on the evolution at $t=1000\tau$. In Fig. 2, we display results for the oscillating potential (25).

The conclusions are essentially the same as with the pulse. Chin and Chen's approximation is more accurate than the other fourth-order approximations with the same number of exponentials of H_0, but less good than algorithm (5). Due to the large number of time steps, fourth-order approximations with time steps around 0.001 present a saturation related to the computer accuracy.

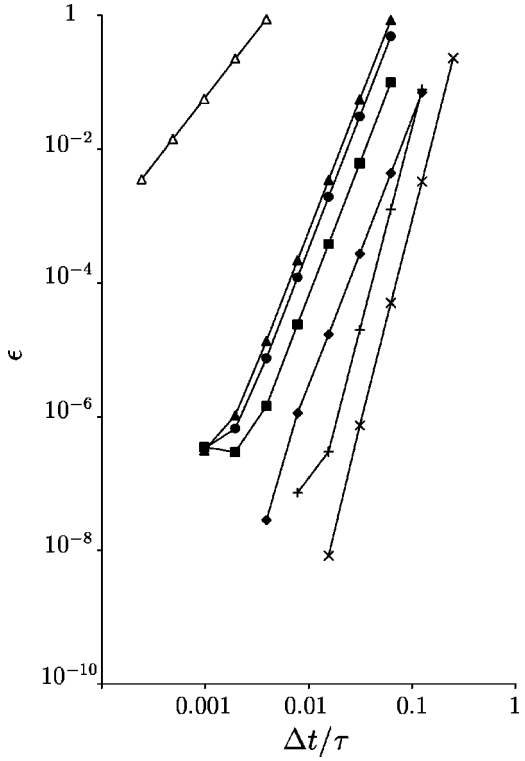

FIG. 2. Accuracies ϵ for the harmonic oscillator forced by oscillating field (26). Second-order factorization (2), open triangles. Fourth-order factorizations (7) with Simpson integration, full triangles; (7) with Gauss integration, full circles; (3) full squares; and (5) full diamonds. Sixth-order factorization (19), crosses, and gradient-free factorization (see text), pluses.

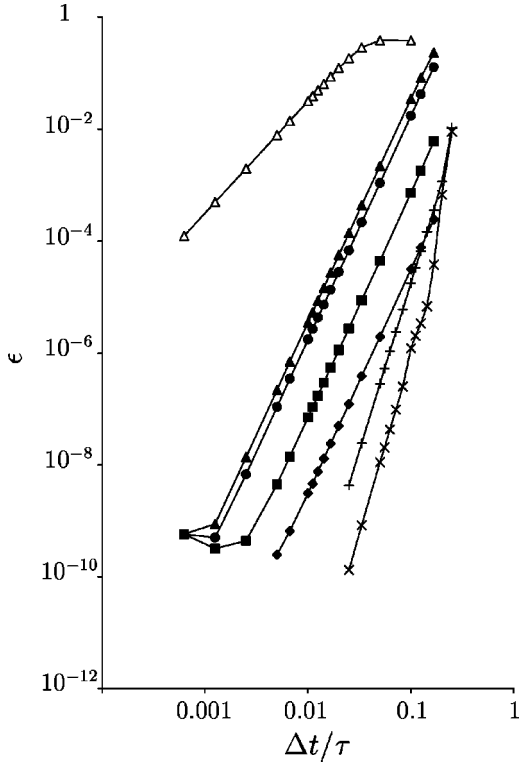

FIG. 3. Accuracies ϵ for the Walker-Preston model. Second-order factorization (2), open triangles. Fourth-order factorizations (7) with Simpson integration, full triangles; (7) with Gauss integration, full circles; (3) full squares; and (5) full diamonds. Sixth-order factorization (19), crosses, and gradient-free factorization (see text), pluses.

The Walker-Preston model [18] is defined by Hamiltonian (21) written as

$$H = -\frac{1}{2m}\frac{\partial^2}{\partial x^2} + V_0(1 - e^{-\alpha x})^2 + Ax\cos\,\omega t, \qquad (28)$$

where $m=1745$, $V_0=0.2251$, $\alpha=1.1741$, $A=0.011025$, and $\omega=0.01787$ [10]. The first two terms represent the standard Morse Hamiltonian H_0 and the last term is the oscillating potential $V(t)$. The wave function is represented on a sinc mesh in the interval $[-0.8, +4.24]$ with $N=80$. The initial wave function is chosen to be the ground state of the time-independent Hamiltonian H_0. As in the previous case, the oscillation period is $\tau=2\pi/\omega$. The final error at $t=1000\tau$ is given by

$$\epsilon = \|\psi^{\text{approx.}}(t) - \psi^{\text{conv.}}(t)\|, \qquad (29)$$

where $\psi^{\text{conv.}}(t)$ is a "converged" wave function obtained at sixth order with small time steps ($\Delta t \leqslant 0.015\tau$). Different approximations of the evolution operator are compared in Fig. 3.

The same behaviors as in the solvable examples are observed. The slopes are about 2 at second order (open triangles), 4 at fourth order (full triangles, full circles, full squares, full diamonds), and 6 at sixth order (crosses, pluses). Again, Chin and Chen's approximation is the best fourth-order algorithm with only two exponentials involving H_0. It seems that it better resists to long-time oscillating fields than the others. The sixth-order algorithm (19) is the most accurate for all time steps.

Since the three examples give consistent information, we discuss the algorithm efficiency in the last case only. In order to compare the computation times of the different approximations, we must take into account the propagation cost of all exponential operators in each factorization. This is estimated with the following assumption: we neglect the computing time of the exponentials of the diagonal matrices representing $V(t)$. This means that the evaluation of this potential at mesh points is considered as negligible. If the potential evaluations were not negligible, the discussion below remains valid if the evaluation time of a pair of two successive exponentials [one with $V(t)$ and one with H_0] is approximately constant. This cannot strictly be true because of the gradient terms. Notice that the present assumptions are quite different from those for classical cases in Ref. [15], so that different conclusions can be expected.

Let n be the average number of exponentials of full H_0 matrices at each time step defined in Sec. II. From Eqs. (2), (3), (7), (5), and (19), n is equal to 1 at second order, 2 or 4 at fourth order, and 5 at sixth order. The gradient-free sixth-order algorithm defined by Eqs. (82) and (83) of Ref. [15] corresponds to $n=7$. In Fig. 4, the total error (29) after the evolution is plotted as a function of $\Delta t/n\tau$, i.e., an effective time step $\Delta t/n$ in τ units.

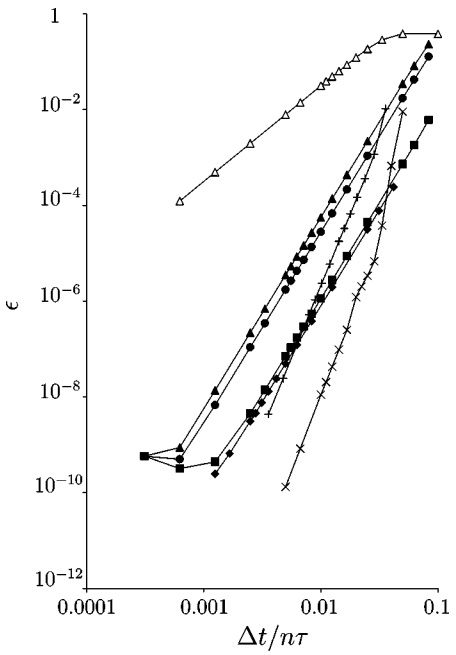

FIG. 4. Accuracies ϵ as a function of the effective time step $\Delta t/n$ (in τ units) for the Walker-Preston model. Second-order factorization (2), open triangles. Fourth-order factorizations (7) with Simpson integration, full triangles; (7) with Gauss integration, full circles; (3) full squares; and (5) full diamonds. Sixth-order factorization (19), crosses, and gradient-free factorization (see text), pluses.

Among the fourth-order algorithms, the simpler algorithm (3) of Chin and Chen becomes competitive with the more complicate algorithm (5). At low accuracy, the sixth-order approximations require more computation time than fourth-order approximations (3) and (5). But at high accuracy, the sixth-order factorizations become faster than all others. However, because of its high number n, the gradient-free algorithm is not competitive. The discussion of the quantum case appears to be quite different from the discussion of the classical case in Ref. [15].

V. CONCLUSION

In this work, we present and discuss a sixth-order algorithm based on the method proposed by Suzuki [9] and on a recent factorization of the exponential of a sum of operators derived in Ref. [14]. Our algorithm of Ref. [11] could not, until now, be extended to sixth order except in the particular case where the gradient term is a constant. Anyway, at fourth order, it is less efficient in the studied examples than the algorithms of Chin and Chen [10] and of Omelyan, Mryglod, and Folk [15], based on the Suzuki procedure. For all time steps, the sixth-order algorithm provides the best accuracy.

Since our main goal is to find the most efficient algorithm for solving time-dependent Schrödinger equations, we estimate the relative efficiency of the algorithms by dividing each time step by a computing-time cost approximated by an integer n. When the required accuracy is not large, fourth-order algorithms and in particular, the simple algorithm of Chin and Chen are the most efficient ones. The sixth-order algorithms are unfavored by the large number of exponentials in approximate factorizations of the evolution operator. However, if a high accuracy is required, the presented sixth-order algorithm (19) becomes the most efficient in our tests and might remain optimal with respect to upper orders.

The efficiency of the different algorithms depends on a subtle balance of accuracy and cost. The definition of efficiency depends on the type of calculation and is quite different for quantal and classical applications. It might also change in calculations more complex than the simple examples discussed here. Anyway, it might be interesting to systematically explore the possible algorithms starting from the classification in Ref. [15].

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APPENDIX

For the pulse defined in Eq. (25), the classical coordinate is given for $0 \leqslant t \leqslant 2$ by

$$x_0(t) = 2K(\pi^2 \cos t - \cos \pi t) + \frac{1}{2}, \tag{A1}$$

with $K = 1/[4(1-\pi^2)]$. The integral of the Lagrangian reads

$$\int_0^t \mathcal{L}_0(t')dt' = -K^2\left(\frac{1+\pi^2}{2\pi}\sin 2\pi t + \pi^4 \sin 2t - 4\pi^2 \sin t \cos \pi t\right) + K\left(\frac{1}{\pi}\sin \pi t - \pi^2 \sin t - \frac{t}{4}\right) - \frac{t}{8}. \tag{A2}$$

For the oscillating field defined in Eq. (26), the classical coordinate is given by

$$x_0(t) = \frac{1}{1-\omega^2}(\sin \omega t - \omega \sin t). \tag{A3}$$

The integral of the Lagrangian reads

$$\int_0^t \mathcal{L}_0(t')dt' = \frac{1}{4(1-\omega^2)^2}\left(\omega^2 \sin 2t + \frac{\omega^2+1}{2\omega}\sin 2\omega t + (\omega^2-1)t - 4\omega \sin \omega t \cos t\right). \tag{A4}$$

In both cases, the initial conditions are $x_0(0)=0$ and $p_0(0)=0$ and the classical momentum is given by $p_0(t)=\dot{x}_0(t)$.

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