Magnus expansion for laser-matter interaction: Application to generic few-cycle laser pulses

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We treat the interaction of an atom with a short intense few-cycle laser pulse by the use of the Magnus expansion of the time-evolution operator. Terms of the Magnus expansion up to the third order in the pulse duration are evaluated explicitly, and expressions for the transition probability between atomic eigenstates are given. The transition operators up to this order are particularly simple, involving only momentum and position shifts arising from the laser field alone. The connection to other existing approaches is established. For a variety of generic short-pulse forms the results of the Magnus approximation are in excellent agreement with time-dependent transition probabilities obtained from accurate *ab initio* numerical calculations. However, the limitation of the Magnus expansion for pulses having both vanishing momentum and position shifts is demonstrated also.

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I. INTRODUCTION

The electronic states of atoms, molecules, and solids can be changed drastically by the interaction with strong laser pulses. Typically large changes (e.g., inversion and ionization) in the occupation probabilities of particular electronic states can be initiated by the strong electric fields. Shaping of the laser pulses allows manipulation of the populations and interesting applications are envisaged, particularly as regard to wave packets generated in the continuum. The designation "strong" is, of course, relative. What is meant roughly is that the field experienced by the electron from the laser is comparable to or greater than the effective atomic electric field (from the nucleus and other electrons) experienced by the electron in its initial state. Since clearly perturbation theory cannot be applied to such pulses, nonperturbative approaches are necessary. In a series of papers [1–4] we have attempted to show that the Magnus expansion [5,6] of the time-evolution operator (TEO) of the laser-atom system provides a very simple but accurate alternative description of these nonlinear processes, provided that the pulse time is short. Again the designation "short" is relative. Roughly it implies that the relevant pulse length is short compared to a classical estimate of the orbital period of an electron in its initial bound state. However, as we shall see, this rough estimate may have to be made more precise depending on the particular form of the pulse considered. Nevertheless, it is clear that for the initial ground states of neutral atoms, attosecond pulses are required but if one prepares initially excited atoms, presently available femtosecond lasers, using the attosecond-pump–femtosecond-probe technique [7,8], are sufficient to bring one into the realm of applicability of the Magnus approximation.

The Magnus expansion leads to a very different picture of ionization in strong short pulses from that obtained from perturbation theory applicable to weak pulses. In the latter, the photon concept is valid, and for long pulses, there is a conservation of energy between the atomic state and the number of photons absorbed. Since this number is small, there is a small transfer of momentum to the atom. In strong short pulses there is no conservation of energy; a broad distribution of ionized electron energies is produced. Furthermore, the momentum transfer to the atom can be large, of the order of a.u., depending on the field strength. As shown in Refs. [1,2], in the first-order Magnus expansion, the key quantity deciding the electron dynamics is the time-dependent momentum transfer $\mathbf{q}(t)$, from the field to the atom, which just corresponds to the instantaneous vector potential, i.e., $\mathbf{q}(t)=\int_0^t dt'\mathbf{E}(t')=-\mathbf{A}(t)$. For pulses consisting of a few half cycles, it is meaningful to follow the electron dynamics over each half cycle. The momentum transfer $\mathbf{q}$ is finite after one half cycle, and if the corresponding energy transfer $(1/2)q^2$ exceeds the ionization potential, then the ionization probability approaches unity and the initial momentum distribution of the electron is transferred as a wave packet to the continuum. Clearly, in a subsequent symmetric half cycle the momentum transfer is reversed and complete recombination should occur since, at the end of a full cycle, $\mathbf{q}=\mathbf{0}$. In this case we showed that it is necessary to go beyond the first-order Magnus, to what we called the modified Magnus approximation (MMA). This involves a consideration of the propagation of the continuum wave packet between the maxima of the half cycles and involves the position shift of the electron during this time.

The dynamics in the strong-field short-pulse limit described by the Magnus expansion also leads to profound differences with the conventional description of strong-field laser-matter interaction (which is usually discussed with reference to a long-pulse laser with wavelength $\lambda=800$ nm). For example, one of the signatures of strong-field short-pulse dynamics is the occurrence of periodic ionization and recombination in time. Namely, we showed that in a sequence of symmetric half cycles, almost complete depletion of the initial state after each odd half cycle can be reversed by the

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subsequent even half cycle of the field. We derived [1,2] closed-form expressions for the recombination probability. Another difference is the movement of the continuum wave packet. Whereas at $\lambda = 800$ nm and for atoms in their ground state only a tiny fraction of the wave packet tunnels out of the nuclear field, is accelerated by the field, and then rescatters from the parent ion; in the strong-field short-pulse limit the *whole* wave packet is transported to the continuum but typically does not move very far from the nucleus. In Refs. [3,4], we showed that such considerations should allow one to manipulate electron wave packets in the continuum even in the case of two-electron atoms.

In a more recent paper [9] we examined in detail the lowest-order terms (in powers of the pulse length) of the Magnus expansion for a half-cycle and a single full-cycle pulse. In particular the gauge invariance of the terms was proven. It also emerged that the momentum $\mathbf{q}$ and position $\boldsymbol{\alpha}$ shifts of the electron by the field are the decisive dynamical factors, and the atomic potential plays a role only in the third-order term. Here we return to this problem and consider the expansion in more detail, apply it to various generic pulse forms, and give explicit examples where the TEO in the lowest orders of the Magnus expansion is compared with the results of an accurate numerical evaluation of the time propagation under the influence of the full TEO. A particular feature of the Magnus expansion is that the lowest-order transition matrix elements between different eigenstates assume very simple and physically intuitive forms. As stated above, in the first order the TEO is simply the momentum boost operator corresponding to a finite momentum transfer $\mathbf{q}$ from the laser to the atom. For pulses where the nett momentum transfer at the end of the pulse is zero, one must proceed to second order, where the transition operator is simply the space translation operator which shifts an initial wave packet to a new location in space [4,9]. Indeed, by employing a generalized "split-operator" strategy we will show here that it is possible to eliminate the effect of the atomic potential from the transition operator up to the third order in the pulse time. Then the atomic potential only occurs in the transition matrix elements through the initial and final eigenstates; the actual time propagation involves properties of the laser pulse only. This leads to a very simple picture of the excitation and recombination processes and gives matrix elements which are quite easy to evaluate but which nevertheless show agreement with numerically accurate results.

In short, the purpose of this paper can be summarized in the following points: to set the limits of applicability and to give a recipe for the calculation of transition probabilities based on a systematic application of the Magnus expansion in powers of the pulse duration, to establish the connection of this approach to existing theoretical methods, and to formulate the Magnus expansion for the calculation of *time-resolved* transition probabilities under the action of short laser pulses.

The plan of this paper is as follows. In Sec. II we describe the Magnus expansion of the TEO both in the Schrödinger and in the interaction pictures and give the explicit form of the expansion up to the third order in the pulse length T. Here the TEO appears as the exponential of a sum of operators. For practical evaluation it is desirable to express this in the simpler form of a product of exponentials, each involving a single operator. This algebraic manipulation is the subject of Sec. III. In Sec. IV, for generic short pulses, we consider the application of the third-order Magnus approximation to calculate various transition probabilities out of an initial hydrogen $1s$ state. The variation in these probabilities as a function of pulse length in the attosecond region is examined. Then in Sec. V the detailed time-dependent variation in transition probabilities during the pulse is calculated. In this section ionization and excitation out of Rydberg states, where the Magnus approximation can be used for pulses of femtosecond duration, is considered also. In all cases the results of the Magnus approximation are compared with the corresponding results of accurate fully numerical calculations of the transition probabilities. Throughout, only the dipole approximation is considered which ultimately limits the magnitude of the field strengths that can be employed and still retain the validity of the Magnus approximation. This is because additional terms involving given powers of the field strength appear in nondipole approximation. All quantities are expressed in a.u.

Several sections are appended to the paper. In Appendix A we discuss the relation of the Magnus expansion to the "sudden-perturbation" expansion [10] which has also been proposed as a way of handling short pulses. In Appendix B the equivalence of the Magnus and perturbation expansions for weak laser fields is established. Also in this appendix, the subtle differences between the present approach and the modified form of the Magnus approximation, the MMA, for few-cycle pulses are exposed. Finally, the details of the algebraic manipulations necessary to derive the results of Sec. III, employing standard forms for the expansion of exponential operators, are given in Appendix C.

II. MAGNUS EXPANSION

We consider the interaction of an atomic or molecular system with a laser pulse in the length gauge and in the dipole approximation. The total Hamiltonian is

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

where H_0 is the Hamiltonian of the field-free atomic/molecular system and

$$H_i(t) = \sum_j \mathbf{r}_j \cdot \mathbf{E}(t) \tag{2}$$

is the time-dependent part of the total Hamiltonian which describes the interaction of the atomic or molecular system with the classical laser field. In the above expression, $\mathbf{r}_j$ is the set of electronic coordinates of the atomic or molecular system and $\mathbf{E}(t)$ is the laser electric field.

We assume that at the onset, before the switch on of the pulse, the system is in some initial state $|\psi(t_0)\rangle = |\phi_i\rangle$. For times $t > t_0$, the state vector $|\psi(t)\rangle$ can then be obtained by application of the TEO to the initial state, i.e.,

$$|\psi(t)\rangle = U(t,t_0)|\phi_i\rangle. \tag{3}$$

The TEO corresponding to the total Hamiltonian $H(t)$ is defined via the time-dependent Schrödinger equation

$$i\partial_t U(t,t_0) = H(t)U(t,t_0), \tag{4}$$

with the initial condition $U(t_0,t_0)=1$.

The solution of Eq. (4) can be expressed formally as $\mathcal{T}\{\exp[-i\int dtH(t)]\}$, where $\mathcal{T}$ is a time-ordering operator. However this formal solution is really simply a prescription of how to expand the exponential as a sum of terms, each corresponding to one order of perturbation theory, i.e., an expansion of the exponential in powers of $H(t)$. Such a perturbation expansion is not applicable to the cases considered here, where the transition probabilities can approach unity. However, in these cases, for pulses which are short compared to the electron orbital times in the atomic or molecular system, one can develop the TEO using the Magnus expansion [5,6]. In the Magnus expansion, with the final time t_e corresponding to the end of the pulse, i.e., $t_e=t_0+T$, the TEO is expressed as an exponential of a sum of terms involving commutators at different times, i.e.,

$$\begin{aligned} U(t_e,t_0) = \exp\Bigg(& -i\int_{t_0}^{t_e} dt' H(t') \\ & + \frac{1}{2}\int_{t_0}^{t_e} dt' \int_{t_0}^{t'} dt''[H(t''),H(t')] \\ & + \frac{i}{6}\int_{t_0}^{t_e} dt' \int_{t_0}^{t'} dt'' \int_{t_0}^{t''} dt'''([H(t'),[H(t''),H(t''')]] \\ & + [[H(t'),H(t'')],H(t''')]) + \cdots \Bigg). \end{aligned} \tag{5}$$

Each term in the above expansion corresponds directly to a certain order in T. However, in this expansion, the terms are *added in the exponent* and not simply added as in the perturbation expansion. Restricting to a certain order of T [truncating the sum in the exponent in Eq. (5)] does not violate the unitarity of the approximate TEO. However since the full Hamiltonian appears in the exponential in Eq. (5) the terms are difficult to evaluate since this Hamiltonian is a sum of kinetic-energy operator, potential-energy operator, and electron-field interaction operator. To make the calculation tractable it is desirable to expand each exponential of a sum of operators into a product of exponentials of a *single* operator. As shown in Ref. [9], this is achieved by splitting the sum in the exponent into a product of exponentials using the Zassenhaus formula [6]

$$\begin{aligned} \exp[(A+B)T] = & \exp[AT]\exp[BT]\exp\{-[A,B]T^2/2\} \\ & \times\exp[[B,[A,B]]T^3/3 + [A,[A,B]]T^3/6] \\ & \times\exp[O(T^4)], \end{aligned} \tag{6}$$

where A and B are two arbitrary operators. Another way is to directly apply the Fer expansion [6] for the TEO,

$$\begin{aligned} U(t_e,t_0) = & \exp\left[-i\int_{t_0}^{t_e} dt' H(t')\right]\exp\left\{\frac{1}{2}\int_{t_0}^{t_e} dt' \int_{t_0}^{t'} dt''[H(t''),H(t')]\right\}\exp\Bigg(\frac{i}{3}\int_{t_0}^{t_e} dt' \int_{t_0}^{t'} dt'' \int_{t_0}^{t''} dt'''\{[H(t''),[H(t'''),H(t')]] \\ & + [H(t'''),[H(t''),H(t')]]\}\Bigg)\ldots, \end{aligned} \tag{7}$$

and thus obtain a product of exponentials. The two methods are essentially equivalent however since if one applies the Zassenhaus formula to Eq. (5) one can put it into the form of expansion (7). One notes that the sudden-perturbation expansion, developed in Ref. [10], as we show explicitly in Appendix A, is clearly different from the Magnus expansion. Unlike the Magnus expansion, it is not unitary and it does not provide a systematic expansion in the powers of the pulse duration.

The Magnus expansion can also be applied to the TEO in the interaction picture, which we denote by $S(t,t_0)$. This TEO also is a solution of the differential Eq. (4), with $U(t,t_0)$ replaced by $S(t,t_0)$ and $H(t)$ replaced by $W(t)=\exp[iH_0t]H_i(t)\exp[-iH_0t]$. Then the Magnus expansion of the TEO in the interaction picture $S(t,t_0)$ is identical to Eq. (5), except that $H(t)$ is replaced by $W(t)$. However unlike expansion (5) in the Schrödinger picture, in the interaction picture, each term in the Magnus expansion does not correspond exactly to a certain order in T. Rather, in general, each term contains an infinite number of orders of T. Therefore, except in the trivial case where H_0 and $H_i(t)$ commute for all times t, a systematic expansion in orders of T by truncating the Magnus expansion in the interaction picture is not possible. Nevertheless, also in the interaction picture, we can evaluate the form of the terms of a given order in T, at least for the lower orders. Hence, for these reasons, in the following when we refer to "order" we will always imply exclusively the order in powers of T and not the Magnus series expansion evaluated to a particular order, since these are not always the same. For all practical purposes, evaluation of the Magnus expansion up to the third order in pulse time is sufficient, since the effort required to evaluate the higher orders becomes equal to or even greater than that of performing a full *ab initio* calculation.

Magnus expansion for laser-matter interaction in the single-active-electron approximation

In the following, we confine ourselves to an atomic system with a single active electron, interacting with a laser

pulse in the dipole approximation. Note that the results are applicable also to molecules that can be described within the single-active-electron approximation. The field-free Hamiltonian is

$$H_0 = \mathbf{p}^2/2 + V, \tag{8}$$

where $\mathbf{p}$ is the momentum operator, $V=V(\mathbf{r})$ is the atomic potential, and $\mathbf{r}$ is the coordinate of the active electron. The time-dependent part of the Hamiltonian is $H_i(t)=\mathbf{r}\cdot\mathbf{E}(t)$.

As shown in Ref. [9], the systematic application of the Magnus expansion in the Schrödinger picture, truncated to a given order in T, is gauge invariant. Then we are free to choose any gauge as long as we consider also the transformation of the initial and final states in the different gauges correctly. For the moment we will use the length gauge. Then the transition matrix elements involve initial and final states which are simply the field-free atomic eigenfunctions. We note that the description of the process in the Kramers-Henneberger frame yields the same transition probabilities, again so long as the transformation factors between the initial and final states in the different representations are included correctly and the expressions in the exponential of the TEO are approximated up to a given order in the pulse length T.

According to Ref. [9], the TEO at time $t_e=t_0+T$ immediately after the action of the pulse, expanded up to the third order in the pulse length T, is given by

$$U(t_e,t_0) = \exp[-iH_0T - i\mathbf{q}\cdot\mathbf{r} - i\mathbf{p}\cdot(\mathbf{q}T/2+\boldsymbol{\alpha}) + i(\boldsymbol{\nabla}V)\cdot(\boldsymbol{\alpha}/2 + \mathbf{q}T/12 - \overline{\boldsymbol{\alpha}})T - i(q^2T/12 + \Phi + \boldsymbol{\alpha}\cdot\mathbf{q}/2) + O(T^4)], \tag{9}$$

with the following definitions:

$$\mathbf{q} = \int_{t_0}^{t_e} dt\mathbf{E}(t) = -\mathbf{A}(t_e), \tag{10}$$

$$\boldsymbol{\alpha} = \int_{t_0}^{t_e} dt\mathbf{A}(t), \tag{11}$$

$$\overline{\boldsymbol{\alpha}} = \frac{1}{T}\int_{t_0}^{t_e} dt' \int_{t_0}^{t'} dt\mathbf{A}(t) = \frac{1}{T}\int_{t_0}^{t_e} dt'\,\boldsymbol{\alpha}(t'), \tag{12}$$

$$\Phi = \int_{t_0}^{t_e} dt'\,\mathbf{A}(t')^2/2, \tag{13}$$

and the laser vector potential $\mathbf{A}(t)=-\int_{t_0}^{t} dt'\mathbf{E}(t')$. Note that each term in Eq. (9) has the dimensions of an action, and in particular $\mathbf{q}$ the dimension of momentum and $\boldsymbol{\alpha}$, $\overline{\boldsymbol{\alpha}}$ of a distance or shift. Each term is of a definite order in T. Specifically, $\mathbf{q}$ is of the first order in T, $\boldsymbol{\alpha}$, and $\overline{\boldsymbol{\alpha}}$, the dimension of distance, of the second order and the unimportant phase Φ of the third order. A close inspection of Eq. (9) reveals that terms of the nth-order Magnus approximation in the Schrödinger picture contains terms up to the nth order in the peak field strength $E_0=\max E(t)$. In the case of weak fields, the TEO of Eq. (9) has a common limit with the first-order time-dependent perturbation theory (see Appendix B, Sec. B 1).

In the interaction picture and in the length gauge the Hamiltonian is

$$W(t) = \exp[iH_0t]\mathbf{E}(t)\cdot\mathbf{r}\exp[-iH_0t]. \tag{14}$$

With the application of the Baker-Hausdorff identity for two operators A and B,

$$\exp[At]B\exp[-At] = B + [A,B]t + [A,[A,B]]\frac{t^2}{2!} + \cdots, \tag{15}$$

the Hamiltonian $W(t)$ can be written as follows:

$$W(t) = [\mathbf{r} + \mathbf{p}t - (\boldsymbol{\nabla}V)t^2/2 + \cdots]\cdot\mathbf{E}(t). \tag{16}$$

This result will be used below when discussing the detailed time dependence of transition probabilities.

We note that in the interaction picture the nth-order Magnus term is proportional to the nth order E_0^n of the peak field strength E_0 but in principle, contains all powers of T. This is to be contrasted with the Schrödinger picture, where the nth-order approximation contains only powers of T^n. Nevertheless, it turns out that the first order in the interaction picture alone contains all terms up to the order T^3. Since, in the following we restrict discussion to terms of this order, the first-order interaction picture also allows a systematic expansion in terms of the pulse duration.

III. EVALUATION OF TRANSITION PROBABILITIES IN THE MAGNUS EXPANSION

The next step is to simplify Eq. (9) to express it as a product of exponentials, each involving a single operator (splitting procedure). Unfortunately, there is no unique procedure for splitting of the TEO of Eq. (9) into a product suitable for evaluation. Except in the cases where we confine ourselves only to the first nonvanishing order, the exact ordering of the operators corresponding to different orders of T is important. This is because different ordering results in different expressions for the transition probability, even though, in the Schrödinger picture, still fulfilling the requirement that the resulting product of exponentials is a TEO of order T^n. In particular it is unclear how to rearrange the factor $\exp[-iH_0T]$, i.e., whether to put it wholly on the extreme left side, extreme right side, or to split it between the left and right sides of the TEO. However, as we shall see, once this is accomplished, this operator then operates directly on the initial or the final state and therefore results only in an unimportant energy phase. We note that in the case of a coherent superposition of eigenstates of H_0 as initial or final state, the factor $\exp[-iH_0T]$ does matter and care must be taken to include the correct energy phase difference. Below we use only eigenstates of H_0 as initial and final states. Extension of the approach presented below to a coherent superposition of states is straightforward and obvious.

A possible strategy to minimize the error in a given order is to split the exponent of a sum of operators such that terms of the order in T that are one order higher than the first

nonvanishing order are eliminated. The elimination of the second order in this way has already been used in the literature (see, e.g., [11]). Here, depending on the pulse properties, we generalize this strategy to eliminate higher orders.

We illustrate the basic idea by looking at the way to split the TEO which contains terms up to second order in T. The truncated expression for the TEO in the Magnus expansion up to the second order is given by

$$U^{(2)}(t_e,t_0) = \exp[-iH_0T - i\mathbf{q}\cdot\mathbf{r} - i\mathbf{p}\cdot(\mathbf{q}T/2 + \boldsymbol{\alpha})]. \quad (17)$$

We introduce a parameter λ such that we are able to adjust the fraction of H_0 in the expression $\exp[-iH_0T]$ which is positioned at the left or the right of the TEO. More precisely, using the Zassenhaus formula (6), the above expression can be transformed into

$$U^{(2)}(t_e,t_0) = \exp[-i\lambda H_0T]\exp[-i\mathbf{q}\cdot\mathbf{r}] \times \exp[-i\mathbf{p}\cdot(\lambda\mathbf{q}T + \boldsymbol{\alpha})]\exp[-i(1-\lambda)H_0T]. \quad (18)$$

It can be seen that with the particular choice

$$\lambda = \lambda^{(2)} \equiv -\boldsymbol{\alpha}\cdot\mathbf{q}/(q^2T), \quad (19)$$

the terms of second order in T are eliminated from the exponent. Thus, while the resulting TEO only contains terms of the first order in T, it is accurate up to the neglect of the third order in T. For pulses with $\mathbf{q}T/2 = -\boldsymbol{\alpha}$, the value $\lambda = 1/2$ is obtained. This important class of pulses includes pulses described by even functions with respect to the center of the pulse, that is, when $E(t_0+T/2-t) = E(t_0+T/2+t)$, for example, the sine half cycle. Note that the TEO which corresponds to this situation is simply

$$U^{(2)}(t_e,t_0) = \exp[-iH_0T/2]\exp[-i\mathbf{q}\cdot\mathbf{r}]\exp[-iH_0T/2]. \quad (20)$$

For all λ the probability of transition between the initial $|\phi_i\rangle$ and final atomic state $|\phi_f\rangle$ is

$$P^{(2)} = |\langle\phi_f|\exp[-i\mathbf{q}\cdot\mathbf{r}]|\phi_i\rangle|^2, \quad (21)$$

where the designation $P^{(2)}$ indicates that the expression is accurate, inclusive of terms of the second order in time, even though the transition operator (evolution operator) is only of the first order.

However this procedure does not work for all pulses since eliminating the second order is not always possible. This can be seen clearly in the case $\mathbf{q}=\mathbf{0}$. Then there is no way to eliminate the second-order term in Eq. (18) since it becomes the leading term in the Magnus expansion. In this case the approximation must be taken up to the third order. Then the following expression is obtained for the TEO, with $\mathbf{q}=\mathbf{0}$,

$$U^{(3)}(t_e,t_0) = \exp[-i\lambda H_0T]\exp[-i\mathbf{p}\cdot\boldsymbol{\alpha}] \times \exp[-i(\boldsymbol{\nabla}V)\cdot(\lambda\boldsymbol{\alpha} - \overline{\boldsymbol{\alpha}})T]\exp[-i(1-\lambda)H_0T]. \quad (22)$$

Now, since the second-order term is the first nonvanishing, it is possible to split in a way which eliminates the third-order term by choosing $\lambda\boldsymbol{\alpha} - \overline{\boldsymbol{\alpha}} = 0$ or

$$\lambda = \lambda^{(3)} \equiv \overline{\boldsymbol{\alpha}}\cdot\boldsymbol{\alpha}/\alpha^2. \quad (23)$$

Then one obtains the transition probability accurate up to the third order given by

$$P^{(3)} = |\langle\phi_f|\exp[-i\mathbf{p}\cdot\boldsymbol{\alpha}]|\phi_i\rangle|^2. \quad (24)$$

Here $\boldsymbol{\alpha}$ is the total displacement of a classical electron during the pulse. This result is valid for all pulses that can propagate, i.e., pulses with $\mathbf{q} = \int_{t_0}^{t_0+T} dt\mathbf{E}(t) = \mathbf{0}$. In the important case of pulses which are odd with respect to the midpoint in time, i.e., $E(t_0+T/2+t) = -E(t_0+T/2-t)$, the optimal choice is $\lambda^{(3)} = 1/2$.

The above splitting strategy becomes insufficient when considering the dynamics *during* the pulse, since now the laser field parameters $\mathbf{q}(t)$ and $\boldsymbol{\alpha}(t)$ are time dependent and are both in general nonzero. Obtaining the TEO in a form where the third-order term in t vanishes would involve the application of the Zassenhaus formula several times and would result in a longish derivation. In this case a better, since algebraic less complex, approach is to use the Magnus expansion in the interaction representation. We confine the discussion to the first term of the Magnus expansion in the interaction picture (as explained above, this is sufficient when including all significant terms up to the third order). To include a parameter that can play the role of λ in the Schrödinger picture, we adopt the convention that the states in the interaction picture $|\Psi_I(t)\rangle$ are defined by

$$|\Psi_I(t)\rangle = \exp[-iH_0(t-t_s)]|\Psi_S(t)\rangle, \quad (25)$$

where $|\Psi_S(t)\rangle$ is the corresponding state in the Schrödinger picture instead of the usual definition $|\Psi_I(t)\rangle = \exp[-iH_0t]|\Psi_S(t)\rangle$. Then the Hamiltonian of Eq. (14) becomes

$$W(t) = \exp[iH_0(t-t_s)]\mathbf{E}(t)\cdot\mathbf{r}\exp[-iH_0(t-t_s)]. \quad (26)$$

The TEO in the interaction picture still satisfies the same differential equation, so that we can apply the Magnus expansion. From Eq. (26) the first term in the Magnus expansion can be written as

$$S(t,0) = \exp\left\{-i\int_0^t [\mathbf{r} + \mathbf{p}(t'-t_s) - (\boldsymbol{\nabla}V)(t'-t_s)^2/2 + \cdots] \cdot \mathbf{E}(t')dt'\right\}. \quad (27)$$

Note that without loss of generality, we assume that all pulses start at time zero. The "virtual" switch-on time t_s of state rotation plays the role of λ in the Schrödinger picture. Namely, t_s induces only an unimportant phase shift ($\exp[\pm iH_0t_s]$) when one considers only eigenstates of the field-free Hamiltonian H_0 as initial and final states. For a coherent superposition of states as initial or final, the correct energy phase must be included explicitly (see the discussion in the first paragraph of this section). Now if we define

$$\mathbf{q}^{(n)}(t) = \frac{1}{(n-1)!}\int_0^t (t'-t_s)^{n-1}\mathbf{E}(t')dt', \tag{28}$$

where n is an integer, noting that $\mathbf{q}^{(n)}$ is of order n in time and further define

$$\mathbf{r}^{(1)} = \mathbf{r} \text{ and } \mathbf{r}^{(i+1)} = [iH_0, \mathbf{r}^i], \quad i = 1,2,\ldots, \tag{29}$$

then the TEO of Eq. (27) can be written as

$$S(t,0) = \exp\left[-i\sum_{n=1}^{\infty}\mathbf{q}^{(n)}(t)\cdot\mathbf{r}^{(n)}\right]. \tag{30}$$

As in the Schrödinger picture, now we can truncate the expression Eq. (30) at a particular order of T and require that the next order is zero. This is possible by adjusting the parameter t_s. To obtain an approximation up to the second order, $\mathbf{q}_2(t)$ should be set to zero, which gives $\mathbf{q}(t-t_s)+\boldsymbol{\alpha}=\mathbf{0}$, so that

$$t_s = t + \frac{\boldsymbol{\alpha}\cdot\mathbf{q}}{q^2} = (1-\lambda^{(2)})t. \tag{31}$$

Hence we have obtained a splitting as in the case of the Schrödinger picture. To obtain the TEO up to the third order, regardless of whether $\mathbf{q}=\mathbf{0}$ or not, we set $\mathbf{q}^{(3)}(t)=\mathbf{0}$ to give a quadratic equation for t_s, i.e.,

$$\frac{1}{2}(t-t_s)^2\mathbf{q} + (t-t_s)\boldsymbol{\alpha} - \overline{\boldsymbol{\alpha}}t = 0. \tag{32}$$

This equation has the solution

$$t_s(t) = t - \frac{-\alpha^2 + \alpha\sqrt{\alpha^2 + 2\overline{\boldsymbol{\alpha}}\cdot\mathbf{q}t}}{\boldsymbol{\alpha}\cdot\mathbf{q}}. \tag{33}$$

For certain pulses and pulse times, the above equation has complex roots which, of course, are unphysical since their inclusion results in a nonunitary TEO. Therefore, it is not possible to set the third-order term to zero for all times t and for all pulses. Then, a more practical approach is simply to take the real part of $t_s(t)$ with the aim of minimizing the third-order correction. Then the transition probability becomes

$$P^{(2)}(t) = |\langle\phi_f|\exp[-i\mathbf{q}^{(1)}(t)\cdot\mathbf{r}]\exp[-i\mathbf{p}\cdot\mathbf{q}^{(2)}(t)]|\phi_i\rangle|^2. \tag{34}$$

Note that in the case $\mathbf{q}=\mathbf{0}$, from either Eq. (32) or (33) one has

$$t_s = t - \frac{\boldsymbol{\alpha}\cdot\overline{\boldsymbol{\alpha}}t}{\alpha^2} = (1-\lambda^{(3)})t, \tag{35}$$

so that the transition probability reduces to Eq. (24) as it should.

To summarize the results of this section, for half-cycle pulses we will use Eq. (21) and for pulses for which $\mathbf{q}=\mathbf{0}$ Eq. (24) will be used. In both cases this gives the transition probability at the end of the pulse. In cases where the actual time dependence of transition probabilities is required, they will be calculated using Eq. (34).

IV. PERFORMANCE OF THE MAGNUS EXPANSION

In the following we compare the results of the Magnus expansion for the interaction of an atom with diverse laser pulses with the results for the same laser pulses obtained from a fully numerical approach. For simplicity and to ensure exact atomic wave functions, consideration is confined to hydrogen as the atomic system. The numerical method used for solving the time-dependent Schrödinger equation is based on an application of the discrete variable representation (DVR) [12]. Since all laser pulses are built up from half-cycle pulses, we begin with a half-cycle pulse and distinguish between symmetric (even functions with respect to their midpoint in time) and asymmetric ones (neither even nor odd functions). The analytic expressions derived for these TEOs can then be used for arbitrary pulses by a consecutive application of TEOs to each half cycle, the sequence of which constitutes the overall pulse. This procedure follows the basic idea of the MMA developed in Ref. [2], where also further approximations are given that lead to the so-called asymptotic MMA. In Appendix B, Sec. B 2 we give the relation of the asymptotic MMA to the second-order Magnus approximation in the Schrödinger picture applied to the total pulse.

Examples of the three generic pulse types for which, (i) $\mathbf{q}\neq\mathbf{0}$ and $\boldsymbol{\alpha}\neq\mathbf{0}$, (ii) $\mathbf{q}=\mathbf{0}$ and $\boldsymbol{\alpha}\neq\mathbf{0}$, and (iii) $\mathbf{q}=\mathbf{0}$ and $\boldsymbol{\alpha}=\mathbf{0}$ will be considered since they require increasingly higher orders in T as the first nonvanishing term. In Appendix B, Sec. B 1 we present the relation of the Magnus approach to the first-order time-dependent perturbation theory for the above pulse classes.

A. Type (i) pulses

The simplest example of type (i), $\mathbf{q}\neq\mathbf{0}$ and $\boldsymbol{\alpha}\neq\mathbf{0}$, are half-cycle pulses with a pulse length $T=\tau$, i.e., τ is the half-cycle time. In Sec. III we have shown that for a pulse with $\mathbf{q}\neq\mathbf{0}$, no matter what the pulse shape, it is possible to split the second-order Magnus result and obtain an expression for the transition probability [Eq. (21)] involving only an operator of the first order in τ but which is accurate inclusive of the second order.

In order to assess the accuracy of the second-order expression, we will consider also the third-order correction to Eq. (21). When we split the TEO of Eq. (9) and use the optimal $\lambda=\lambda^{(2)}$ of Eq. (19) we obtain

$$\begin{aligned} U^{(3)}(\tau,0) = \exp[-i\lambda H_0\tau]\exp\{-i\mathbf{q}\cdot\mathbf{r} + i(\boldsymbol{\nabla}V)\cdot[\boldsymbol{\alpha} + (\lambda^2/2 \\ + 1/4)\mathbf{q}\tau - \overline{\boldsymbol{\alpha}}]\tau\}\exp[-i(\Phi + \boldsymbol{\alpha}\cdot\mathbf{q}/2)] \\ \times\exp[-i(1-\lambda)H_0\tau]\exp[O(\tau^4)]. \end{aligned} \tag{36}$$

The derivation of this equation is given in Appendix C.

The probability of a transition between the initial $|\phi_i\rangle$ and final atomic state $|\phi_f\rangle$ for a general "asymmetric" half-cycle pulse in the third order in τ is

$$\begin{aligned} P^{(3)} = |\langle\phi_f|\exp\{-i\mathbf{q}\cdot\mathbf{r} + i(\boldsymbol{\nabla}V)\cdot[\boldsymbol{\alpha} + (\lambda^2/2 + 1/4)\mathbf{q}\tau - \overline{\boldsymbol{\alpha}}]\tau\} \\ \times|\phi_i\rangle|^2. \end{aligned} \tag{37}$$

Note that the operator in this expression is much simpler

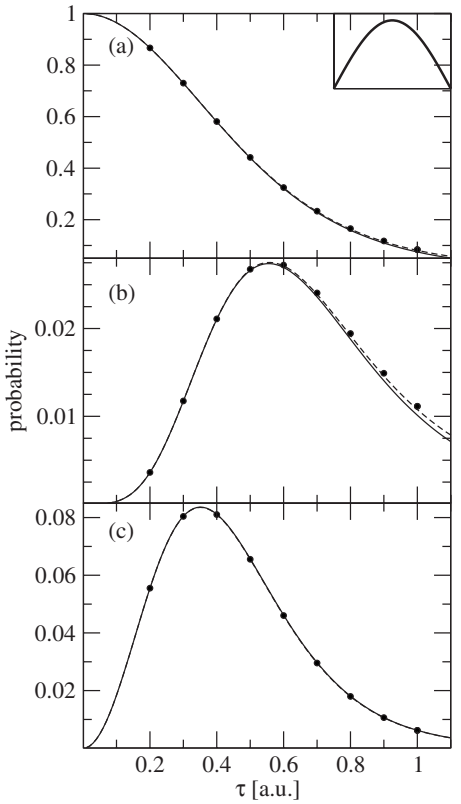

FIG. 1. (a) Occupation probability of the initial $1s$ state, (b) excitation probability to the $2s$ state, and (c) to the $2p$ state following a half-cycle sine pulse with $E_0=3$ a.u. as a function of τ. The results are as follows: circles, numerical; solid line, first-order Magnus of Eq. (21); dashed line, third-order Magnus of Eq. (39).

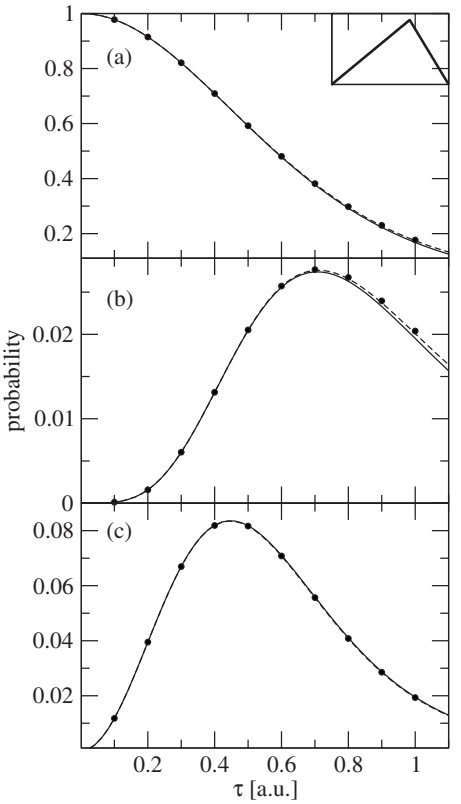

FIG. 2. (a) Occupation probability of the initial $1s$ state, (b) excitation probability to the $2s$ state and (c) to the $2p$ state following a half-cycle triangular pulse with $E_0=3$ a.u. as a function of τ. The results are as follows: circles, numerical; solid line, first-order Magnus of Eq. (21); dashed line, third-order Magnus of Eq. (37).

than that of Eq. (36), since trivial phase factors cancel from the probability. In the special case of a symmetric half-cycle pulse (even function in time), the condition $\boldsymbol{\alpha}=-\mathbf{q}\tau/2$ between the momentum kick and the electron displacement is valid, i.e., the splitting is symmetric ($\lambda=1/2$) and Eq. (36) takes the simpler form

$$U^{(3)}(\tau,0)=\exp[-iH_0\tau/2]\exp[-i\mathbf{q}\cdot\mathbf{r}-i(\boldsymbol{\nabla}V)\cdot(\mathbf{q}\tau/8+\overline{\boldsymbol{\alpha}})\tau]\exp[i(q^2\tau/4-\Phi)]\exp[-iH_0\tau/2]. \quad (38)$$

The third-order transition probability then becomes

$$P^{(3)}=|\langle\phi_f|\exp[-i\mathbf{q}\cdot\mathbf{r}-i(\boldsymbol{\nabla}V)\cdot(\mathbf{q}\tau/8+\overline{\boldsymbol{\alpha}})\tau]|\phi_i\rangle|^2. \quad (39)$$

By comparing Eqs. (37) and (39), one sees that the asymmetry of the half-cycle pulse only affects the third-order term.

In Figs. 1 and 2 we show the occupation probability of the ground state and the excitation probability into the $2s$ and $2p$ states, after the interaction with a (symmetric) half-cycle sine pulse and with an asymmetric half-cycle pulse, for various pulse lengths. The sine half-cycle pulse is

$$\mathbf{E}(t)=\mathbf{E}_0\sin\left(\frac{\pi t}{\tau}\right),\quad 0\le t\le\tau, \quad (40)$$

whereas for simplicity, although unphysical, we take the asymmetric pulse to have a triangular shape,

$$\mathbf{E}(t)=\mathbf{E}_0\begin{cases}3t/(2\tau), & 0<t<2\tau/3\\ 3\left(1-\frac{t}{\tau}\right), & 2\tau/3<t<\tau\\ 0, & \text{otherwise},\end{cases} \quad (41)$$

whose form is given in the inset in the top right-hand corner of Fig. 2. The comparison with the numerical calculations reveals that Eq. (21) in both cases is very good and that the third-order corrections of Eqs. (39) Fig. 1) and (37) (Fig. 2) are minor. However, the corrections are noticeable for the larger pulse lengths, $\tau>0.5$ a.u. For such pulse lengths the third-order prediction improves the agreement with the numerical data.

The probabilities illustrate the pulse-length τ dependence (which translates into momentum-transfer q dependence) observed in Ref. [2]. As τ increases the population transfers to excited and ionized states, but, as shown by the subsequent decline of the excited-state probabilities, at the end of the half cycle when τ approaches unity, almost 80% of the population resides in the continuum. One notes also the good accuracy of the Magnus results up to pulse lengths equal to the initial electron orbital time, which is beyond what one might expect.

In Fig. 3 a cut through the momentum distribution $dP_{\text{ion}}/d^3\mathbf{k}$ of the ionized electrons is shown. The momentum component is in the laser polarization direction [$\mathbf{k}$

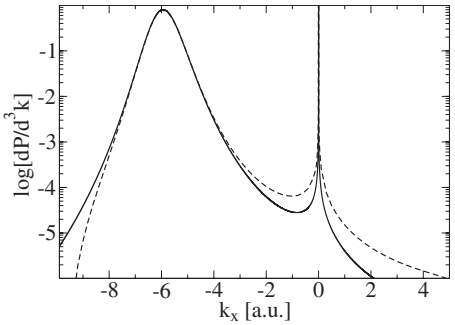

FIG. 3. Ionization probability from the initial $1s$ state into continuum states $|\psi_{\mathbf{k}}\rangle$ with momentum $\mathbf{k}=(k,0,0)$ following a half-cycle sine pulse with $\tau=0.5$ a.u. and $E_0=6\pi$ a.u. Solid line, first-order Magnus; dashed line, third order. The base of the logarithm is 10.

$=(k,0,0)]$, and ionization is due to a strong half-cycle sine pulse of electric field strength $E_0=6\pi$ a.u. and pulse duration $\tau=0.5$ a.u. This is equivalent to a momentum kick by the laser pulse of $q=6$ a.u. This momentum transfer is evident in the momentum distribution as a peak approximately at the electron momentum $\mathbf{k}=-\mathbf{q}$. Differences between first- and third-order Magnus approximations become visible only on a logarithmic plot. The strong peak at zero momentum is due to the singularity in the Coulomb density of states arising from the attractive atomic potential.

In summary, we conclude that a consideration only of the leading first order in the Magnus expansion, represented simply as a momentum "kick" from the laser pulse, is sufficient to describe the action of a half-cycle pulse and is independent of the pulse shape. The second-order term can be eliminated. The third-order term does depend on the pulse shape but only needs to be taken into account when pulse lengths become comparable to the electronic orbital time. This concludes the discussion of type (i) pulses.

B. Type (ii) pulses

This case applies to pulses such that $\mathbf{q}=\mathbf{0}$ and $\boldsymbol{\alpha}\neq\mathbf{0}$. We consider one-cycle pulses with duration $T=2\tau$ as typical examples of type (ii). In these cases the transition probability can be approximated by the third-order expression in Eq. (24). One-cycle pulses which are odd functions with respect to their midpoint clearly satisfy the condition $\mathbf{q}=\mathbf{0}$. We will consider two separate examples, which are the one-cycle extensions of the half-cycle forms of Sec. IV A. In both cases one can use Eq. (24) to approximate the exact numerical results. We use these cases also to illustrate the difference of the MMA of Ref. [2] and the present approach of Eq. (24) (see also Appendix B).

As a first example, we take the one-cycle sine pulse of the form of Eq. (40) with pulse duration $T=2\tau$. For such a pulse we have $\mathbf{q}=\mathbf{0}$ and $\boldsymbol{\alpha}=-\mathbf{q}_{\rm hc}\tau$. Here $\mathbf{q}_{\rm hc}$ is the half-cycle momentum kick, and Eq. (24) can be written as

$$P^{(3)}=|\langle\phi_f|\exp[i\mathbf{q}_{\rm hc}\cdot\mathbf{p}\tau]|\phi_i\rangle|^2, \tag{42}$$

an expression which is also used in Refs. [1,2,13].

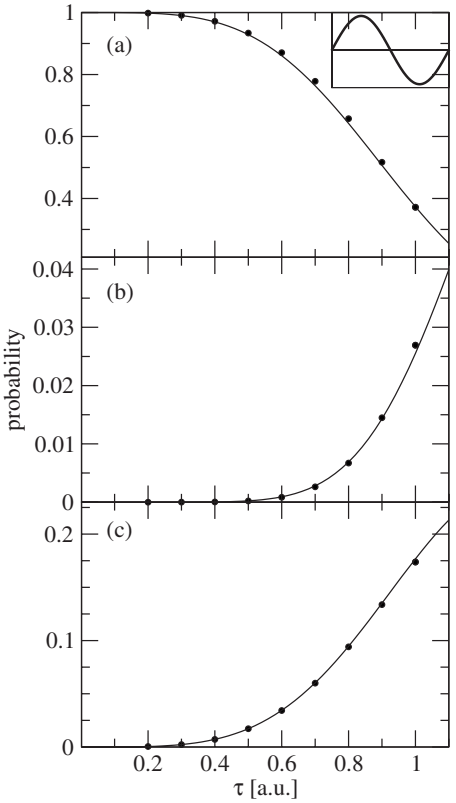

FIG. 4. (a) Occupation probability of the initial $1s$ state, (b) excitation probability to the $2s$ state, and (c) to the $2p$ state following a full-cycle sine pulse with $E_0=3$ a.u. as a function of τ. The results are as follows: circles, numerical; solid line, Eq. (24) or equivalently Eq. (42).

The second example is a full-cycle pulse composed of two identical triangular-shaped half cycles of Eq. (41) but oppositely directed and put together so that the full-cycle pulse is an odd function of time with respect to the midpoint. This pulse is defined by

$$\mathbf{E}(t)=-\mathbf{E}_0\begin{cases}3t/\tau, & |t|<\tau/3\\ \frac{3}{2}(-|t|/\tau+1), & \tau/3<|t|<\tau\\ 0, & \text{otherwise},\end{cases} \tag{43}$$

as shown in the inset of Fig. 5. The probability of transition is calculated using Eq. (24). However, the displacement of the classical electron $\boldsymbol{\alpha}$ can be rewritten in a form where the momentum transfer of the half cycle $\mathbf{q}_{\rm hc}$ appears. This yields $\boldsymbol{\alpha}=-2(1-\lambda_{\rm hc})\mathbf{q}_{\rm hc}\tau$. Therefore the transition probability becomes

$$P^{(3)}=|\langle\phi_f|\exp[2i(1-\lambda_{\rm hc})\mathbf{q}_{\rm hc}\cdot\mathbf{p}\tau]|\phi_i\rangle|^2, \tag{44}$$

with $\lambda_{\rm hc}=\lambda^{(2)}$ for the triangle-shaped half cycle.

We illustrate the performance of the Magnus expansion in both cases in Figs. 4 and 5. In Fig. 4 we show the occupation probability into the initial $1s$ ground state and the excitation probability into the $2s$ and $2p$ states after the action of a full-cycle sine pulse. In Fig. 5 we show the same probabilities following the action of a full-cycle pulse consisting of

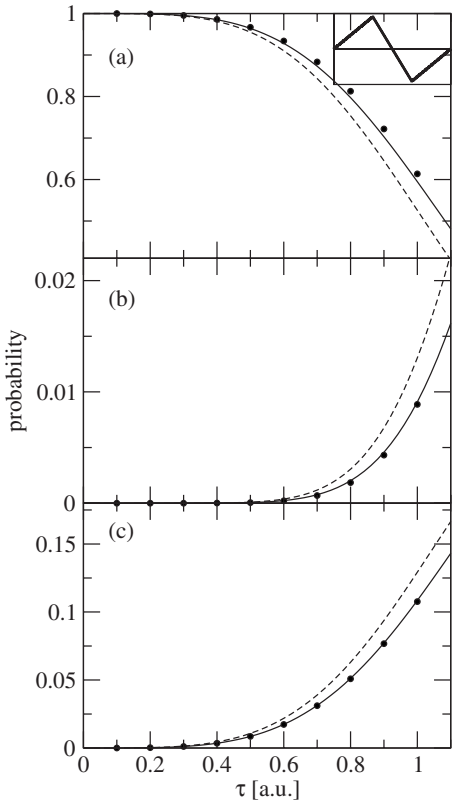

FIG. 5. (a) Occupation probability of the initial $1s$ state, (b) excitation probability to the $2s$ state, and (c) to the $2p$ state following the full-cycle pulse shown in the inset, with $E_0=3$ a.u., as a function of τ. The results are as follows: circles, numerical; solid line, Eq. (44) or equivalently Eq. (24); dashed line, Eq. (42).

the asymmetric triangular half cycles. The probabilities are shown as a function of half-cycle pulse length τ.

For pulses without envelope, that is, pulses consisting of only symmetric half cycles, the results from Eq. (42) and the MMA are identical (see also Appendix B). To illustrate the importance of using Eq. (44) for asymmetric pulses, rather than the MMA, in Fig. 5 we show as dashed line the results obtained using Eq. (42). Clearly better agreement with numerical values is obtained with Eq. (44). Note that in our previous applications of the MMA [1,2] only symmetric half cycles were considered so that the use of Eq. (42) was justified.

In both cases, for small τ, in the first half cycle of the field substantial depletion of the initial state (see also Figs. 1 and 2) and thus ionization occurs, and in the next half cycle of the field the process is reversed so that almost the whole population comes back (recombines) in the initial state. For example, the recombination probability is still 0.9 when τ has reached 0.5 a.u. Such periodic occurrence of ionization and recombination is a signature of the short-pulse strong-field limit and was analyzed in Ref. [1] already. As τ increases the initial state becomes depleted. The momentum transfer of the first half cycle decides the extent of the depletion (the larger the momentum transfer, the larger the depletion). To summarize, as evident in the cases presented in Figs. 4 and 5, the extent of recombination is decided by the half-cycle duration τ (smaller τ's result in higher recombination). For small pulse length the initial-state occupation probability in both cases is almost unity. Not until $\tau=0.3$ a.u., at a maximal laser electric field strength of $E_0=3$ a.u., are some of the electron populations transferred into the energetically higher states. The third-order Magnus transition probability of Eq. (24) reproduces the numerical data well, again, even up to half-cycle pulse lengths of $\tau=1$ a.u., which is the orbital time in the initial state (note the total pulse duration $\tau=2$ a.u.). The decline of the final occupation probability of the initial state (in fact, the recombination probability with respect to the second half cycle) with pulse length is explained by the increase in the position shift $\boldsymbol{\alpha}$, which leads to a lower overlap of the final wave packet with the initial state. Again the population of excited states remains small, indicating that at the field strength of 3 a.u. most probability depleted from the initial state is in the continuum. These results demonstrate the accuracy of the leading-order expression in the Magnus expansion for a full-cycle pulse with zero-momentum kick.

Finally in this section we investigate if the Magnus expansion, Eq. (24), also gives good results for pulses of type (ii) that have a time-varying envelope. Specifically, we consider a two-cycle pulse with Gaussian envelope as follows:

$$\mathbf{E}(t) = \mathbf{E}_0 \exp[-t^2/\tau^2]\sin[\pi t/\tau], \quad |t| < 2\tau, \tag{45}$$

where the half-cycle duration is τ, the total pulse duration is $T=4\tau$, and the maximal electric field strength is $E_0=3.6$ a.u., see the inset of Fig. 6.

In Fig. 6 we display the occupation probability of the initial $1s$ state and excitation probabilities into the $2s$ and $2p$ states following the action of the two-cycle pulse of Eq. (45). The performance of the third-order approximation (24) is excellent until approximately half-cycle pulse lengths of $\tau \sim 0.5$ a.u. and then decreases. The reason for the error (although only at the few percent level) is the neglect of continuum wave-packet spreading. This effect starts to play a role at the larger total pulse lengths of $T=4\tau$. Note, however, that the total pulse length is then much larger than the orbital time 1 a.u. in the initial state.

C. Type (iii) pulses

The final case, that of pulses with

$$\mathbf{q} = \mathbf{0}, \quad \boldsymbol{\alpha} = \mathbf{0}, \quad \text{and} \quad \overline{\boldsymbol{\alpha}} \neq \mathbf{0}, \tag{46}$$

is, as we will see, a special case in which the expansion to third order is tested severely. Note however that for pulses which can propagate, only the condition $\mathbf{q}=\mathbf{0}$ is necessary at the end of the pulse and not the more stringent condition Eq. (46), see, e.g., Ref. [14]. We give these results here simply for the sake of assessment of the performance of the Magnus expansion and less because it has been argued that stabilization of atomic ionization occurs for such pulses [15].

In this case, the leading term in the Magnus expansion is the third-order term in pulse duration, represented by the operator

$$U^{(3)}(T,0) = \exp[-iH_0T - i(\boldsymbol{\nabla} V)\cdot\overline{\boldsymbol{\alpha}}T], \tag{47}$$

where we have omitted the term $-i\Phi$ which is present [see Eq. (9)] but results in an unimportant phase. The term with

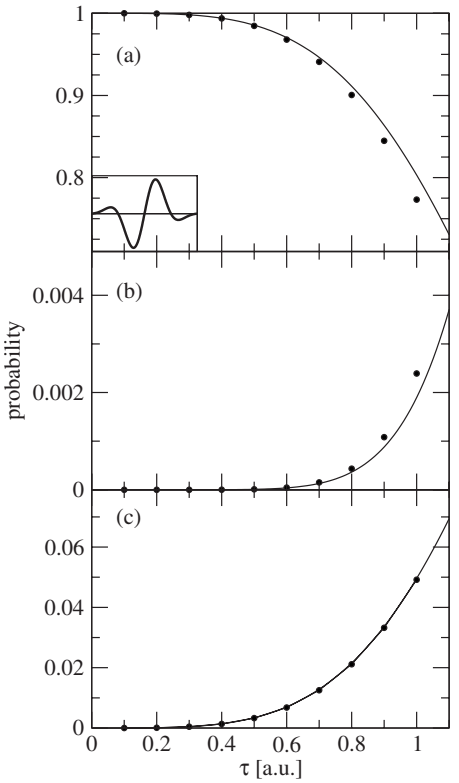

FIG. 6. (a) Occupation probability of the initial $1s$ state, (b) excitation probability to the $2s$ state, and (c) to the $2p$ state following the two-cycle pulse shown in the inset, with $E_0=3.6$ a.u., as a function of τ. The results are as follows: circles, numerical; solid line, Eq. (24).

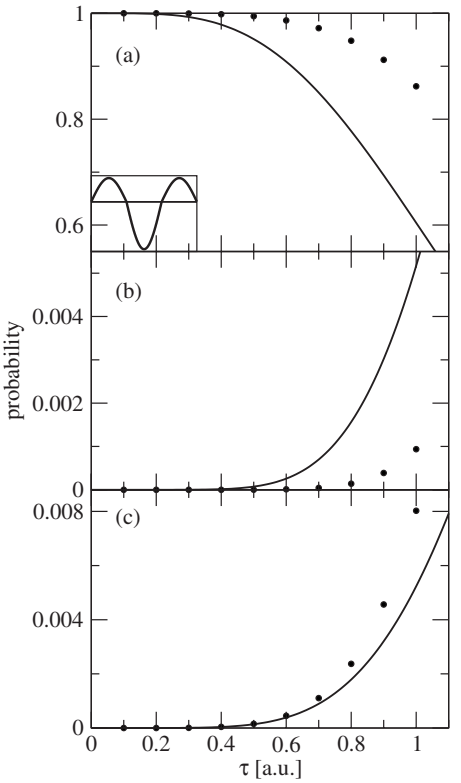

FIG. 7. (a) Occupation probability of the initial $1s$ state, (b) excitation probability to the $2s$ state, and (c) to the $2p$ state following the pulse shown in the inset, with $E_0=3$ a.u., as a function of τ. The results are as follows: circles, numerical; solid line, Eq. (48).

$(\nabla V)\cdot\bar{\boldsymbol{\alpha}}T$ appearing in Eq. (47) is eliminated in the optimal spitting procedure when deriving Eq. (24). However, when $\boldsymbol{\alpha}$ becomes zero, which implies that the displacement of the classical electron induced by the field becomes zero, it is impossible to find an optimal $\lambda\equiv\lambda^{(3)}$ since the third-order term in pulse duration in the Magnus expansion becomes the leading one. Splitting operator (47) the transition probability assumes the form

$$P=|\langle\phi_f|\exp[-i(\nabla V)\cdot\bar{\boldsymbol{\alpha}}T]|\phi_i\rangle|^2. \tag{48}$$

To illustrate the performance of the above expression we use the simplest possible pulse which satisfies condition (46). This consists of three half cycles given by

$$\mathbf{E}(t)=\mathbf{E}_0\begin{cases}(1/2)\sin(\pi t/\tau) & 0<t<\tau\\ \sin(\pi t/\tau) & \tau<t<2\tau\\ (1/2)\sin(\pi t/\tau) & 2\tau<t<3\tau,\end{cases} \tag{49}$$

as shown in the inset of Fig. 7. In Fig. 7 we show the survival probability of the ground state along with the transition probability to the first two excited states, $2s$ and $2p$.

Note that condition (46) in Eq. (24) would imply that probability resides wholly in the initial state at the end of the pulse. As one sees from Fig. 7, up to the accuracy of the plot, this is indeed the result of the numerical calculation for times up to $\tau\sim 0.3$. Hence, up to these pulse lengths one can infer that Eq. (24) holds. However, for longer times one sees that the corrected Eq. (48) gives only a rough qualitative description of the accurate transition probabilities. Since in these cases the third-order approximation is not good, one might be tempted to push the expansion to higher order. However, the fourth-order term can be eliminated by applying the optimal splitting to TEO (47) so that the first nonzero correction term is of the fifth order in pulse duration. The effort required to calculate the fifth-order term in pulse duration is comparable to the full numerical solution of the TEO, and we will not evaluate it.

Although, for pulses satisfying condition (46), it appears that the final transition probabilities are accurate for times up to $\tau\sim 0.3$, it remains to examine whether the detailed time dependence during the pulse is described accurately. This will be done in Sec. V.

V. MAGNUS APPROXIMATION FOR TIME-RESOLVED TRANSITION PROBABILITIES

A more sensitive test of the theory than the dependence of the transition probabilities on total pulse length, i.e., at the end of the pulse, is the detailed time dependence during the pulse. We have shown that in the cases presented here, beginning in the ground $1s$ state, the transition probability at the end of the pulse predicted by numerical calculation is also given accurately by the appropriate lowest-order Magnus approximation (except of course in Fig. 7 for $\tau\geq 0.3$ a.u.). In this section we compare the numerically ob-

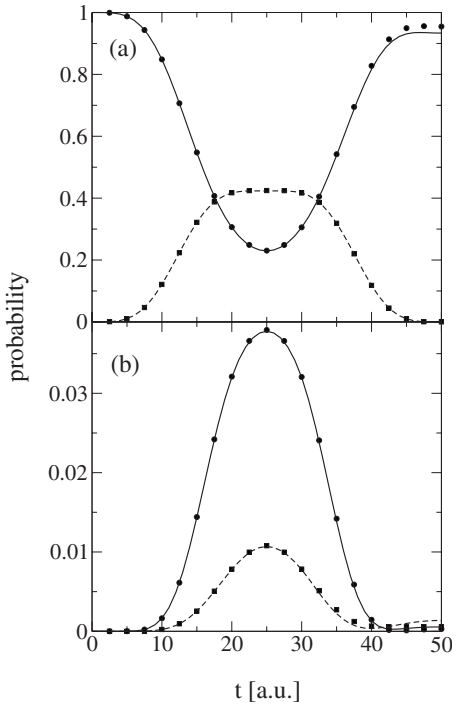

FIG. 8. (a) Time dependence of the occupation probability of the $4s$ (solid line) and $4p$ (dashed line) states and (b) of the $5s$ (solid line) and $3s$ (dashed line) states. In all cases points are the numerical, and lines are the Magnus approximation results. A full-cycle sine pulse with $\tau=25$ a.u. and $E_0=0.005$ a.u. is used.

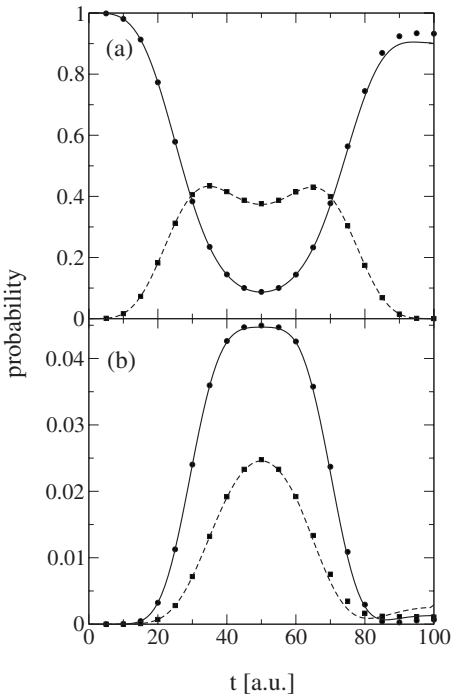

FIG. 9. (a) Time dependence of the occupation probability of the $5s$ (solid line) and $5p$ (dashed line) states and (b) of the $6s$ (solid line) and $4s$ (dashed line) states. In all cases points are the numerical, and lines are the Magnus approximation results. A full-cycle sine pulse with $\tau=50$ a.u. and $E_0=0.002$ a.u. is used.

tained time-resolved probabilities with those predicted by the lowest-order Magnus approximation. Here, to emphasize the applicability of the Magnus expansion also to femtosecond pulses, we will consider the ionization of initial Rydberg states. With the advent of attosecond-femtosecond pump-probe experiments [7,8] it is possible to monitor the time dependence of excitation and ionization probabilities [16]. We imagine an experiment where $H(1s)$ is excited by an attosecond pulse to the fourth or fifth shell, and then a visible laser pulse is applied to the Rydberg state as initial state.

When strong and short pulses with wavelengths in the visible range $\lambda=400-800$ nm (which corresponds to a half-cycle pulse duration of $\tau\approx 25-50$ a.u. or roughly a femtosecond) act on atoms in their *ground* state, the ionization process can be described as multiphoton absorption or as tunneling, depending on the binding energy and the laser intensity. This is because a half-cycle duration of approximately 50 a.u. is much longer than the typical orbital times of outer electrons in their ground state, which is of the order of 1 a.u or less. However, for the *excited* states of hydrogen, say with $n\geq 4$, this half-cycle duration is comparable with the orbital times. Therefore, in these cases the Magnus approximation should apply. However, since for visible light the half-cycle duration is not shorter than the orbital times in low-lying states, one could argue that transitions from the states with $n=4$ and 5 to these lower-lying states is not well described by the Magnus approach. However, as we now demonstrate, the Magnus approximation gives a good description of these transitions also.

We consider that after an $H(1s)$ electron has been excited into the fourth or fifth shell, a visible laser pulse is applied to excite or ionize the electron. This pulse is taken as a one-cycle sine pulse of Eq. (40) [i.e., of type (ii)] of a half-cycle duration of 25 a.u. (wavelength of 362 nm for $n=4$) or 50 a.u. (wavelength of 725 nm for $n=5$). To obtain the time-resolved transition probability from excited states we use Eq. (34) together with $t_s(t)$ taken as the real part of the expression in Eq. (33). A full numerical calculation of the time-dependent wave function evolving from the excited $4s$ and $5s$ states has been performed also, from which numerically accurate transition probabilities can be extracted. We compare the results of the Magnus approximation (34) to the numerical results for (a) the occupation probability of the initial s state and the excitation probability into the p-type state of the same shell and (b) the excitation probabilities into the next energetically higher and lower bound s states, following a full-cycle sine pulse. The transition probabilities as a function of time for such full-cycle pulses are displayed in Figs. 8 and 9. The results of Eq. (34) are in excellent agreement with the numerical ones even for transitions into bound states lower than the initial state. Note that the fourth (fifth) shell is the lowest shell for which the Magnus approach can be applied for 400-nm (800-nm) pulses, since for lower shells the orbital time is too short. By the same token, for higher-lying Rydberg states as initial state, the Magnus approximation should be even more accurate.

Figures 8 and 9 illustrate the typical dependence on time during the pulse, as seen in Ref. [2] for the initial $1s$ and $2s$ states. There is a strong depletion of the initial state and concomitant population of excited states after the first half cycle, followed by a strong reversal of transition at full cycle,

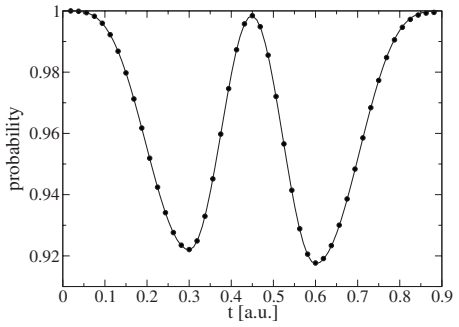

FIG. 10. Time dependence of the $1s$ occupation probability under the action of pulse (49) with $E_0=3$ a.u. and $\tau=0.3$ a.u. Results are as follows: circles, numerical; solid line, Magnus expansion result of Eq. (34).

as expected for $\mathbf{q}=\mathbf{0}$. Again the population of p states dominates those of s states.

Finally, in Fig. 10 we compare the time-dependent Magnus transition probability (34) to the exact numerical results for the case of a type (iii) pulse, specifically that shown in Fig. 7 for $E_0=3.0$ a.u. and $\tau=0.3$ a.u. It can be seen that although at the end of the pulse the survival probability is practically unity, *during* the pulse the population of the initial state oscillates as a function of time. In this case also, the time-dependent Magnus expression (34) agrees excellently with the numerical results. The smallness of the transition probability out of the $1s$ state is presumably due to the fact that the $|\mathbf{q}|$ value of the first half cycle is only approximately 0.3 a.u. so that $\frac{1}{2}\mathbf{q}^2$ is well below the ionization energy.

VI. CONCLUSIONS

In this paper we have examined the first three orders of the expansion of the Magnus approximation to the TEO for propagation in a strong laser field. The expansion parameter is the pulse time T, measured in units of the initial-state classical orbital time. A splitting strategy has been introduced with which the term of order higher than the first nonvanishing one can be eliminated or made small. In the first order this strategy (accurate then to second order) gives transition probabilities at the end of the pulse which depend only on the total momentum transfer $\mathbf{q}$ from the field to the atom. Where this is zero, the second-order (accurate to third order) expression involves the total position shift $\boldsymbol{\alpha}$ of the electron under the acceleration due to the field. Where both $\mathbf{q}$ and $\boldsymbol{\alpha}$ are zero, the third-order term must be used, which involves not only the mean position shift by the field but also the gradient of the atomic potential. Probabilities calculated with these formulae applied to appropriate generic pulses, consisting of up to four half cycles, have been compared to the results of numerically accurate calculations. For the first two types of pulse, excellent agreement is found in all cases, even up to times which are comparable to, or even greater than, the orbital time. Here it appears that only the half-cycle time, not the total pulse time, needs to be short to ensure accuracy of the lowest-order Magnus expression. This result, we feel, is important since it extends the validity of the Magnus approximation, although how strongly the accuracy degenerates as the number of cycles increases is not clear. In the case of type (iii) pulses, where the first two orders of the TEO become unity, agreement with numerical results is limited to short half-cycle times and the Magnus expression breaks down for times approaching the orbital time.

The splitting strategy has also been generalized to calculate transition probabilities not only at the end of the pulse but also for all times during the pulse. Again very good agreement with numerical results is obtained.

The Magnus expansion, as presented here up to third order, gives easily evaluated physically intuitive simple forms for transition operators and probabilities which mimic the dynamics in the short-pulse strong-field limit. The theory is gauge invariant and unitary, making it particularly suitable for treating strong short laser pulses (able to considerably deplete the initial state) as will be encountered in attosecond laser experiments, seeking to explore the real-time dynamics of atomic electrons. Nevertheless, one outstanding problem is the need to delineate the applicability of the results presented here as the laser field strength is increased. This will involve a full consideration of nondipole and relativistic effects.

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APPENDIX A: RELATION TO THE SUDDEN-PERTURBATION EXPANSION

In the following we compare expansions (5)–(7) to the sudden-perturbation expansion of Ref. [10]. Such an expansion is possible when $[H_i(t),H_i(t')]=0$ for all times t and t'. The sudden-perturbation expansion has been formulated in the interaction picture. Then the TEO $S(t_e,t_0)$ is expanded as (Eq. (17) in Ref. [10])

$$S(t_e,t_0)=S_0(t_e,t_0)[1+\Delta_1(t_e,t_0)+\Delta_2(t_e,t_0)+\cdots], \quad \text{(A1)}$$

where

$$S_0(t_e,t')=\exp\left[-i\int_{t_0}^{t_e}dt'W_0(t')\right],$$

$$\Delta_1(t_e,t_0)=-i\int_{t_0}^{t_e}dt'S_0^{-1}(t',t_0)W_1(t')S_0(t',t_0),$$

$$\Delta_2(t_e,t_0)=\frac{\Delta_1^2(t_e,t_0)}{2}-i\int_{t_0}^{t_e}dt'S_0^{-1}(t',t_0)W_2(t')S_0(t',t_0), \quad \text{(A2)}$$

and

$$W_0(t)=\exp[iH_0t_0]H_i(t)\exp[-iH_0t_0],$$

$$W_1(t)=-i(t-t_0)\exp[iH_0t_0][H_i(t),H_0]\exp[-iH_0t_0], \quad \text{(A3)}$$

$$W_2(t) = -\frac{(t-t_0)^2}{2}\exp[iH_0t_0][[H_i(t),H_0],H_0]\exp[-iH_0t_0].$$

The terms $\Delta_n \sim (\omega T)^n$, where ω is "a typical eigenvalue" of H_0 [10].

It is immediately clear that the successive terms in the expansion are *added*, rather then *multiplied*, as in the case with expansion (7). Therefore, in contrast to the Magnus expansion, where by truncating to any number of terms unitarity is preserved, the sudden-perturbation expansion is nonunitary. The only exception is the first term which, up to an unimportant phase factor, is identical to the first term in the Magnus and Fer expansion in the Schrödinger picture. It is the unitarity of the Magnus expansion which makes it suitable for the description of strong perturbations.

APPENDIX B

1. Relation to the time-dependent perturbation theory

The time-dependent perturbation series, obtained by iterating the integral equation corresponding to Eq. (4), is clearly different from the Magnus expansion. However, in the limit of weak and short interactions the two approximations have the same form. Here we derive this common limiting form of first-order perturbation theory and the lowest nonvanishing order of the Magnus expansion for different types of laser pulse.

The first case we investigate are pulses where the overall momentum kick of the pulse is $\mathbf{q} \neq \mathbf{0}$. The transition probability from the initial ground state into a final state in the first-order perturbation theory is then

$$P = \left| -i\int_{t_0}^{t_e} dt\langle\phi_f|\mathbf{r}\cdot\mathbf{E}(t)|\phi_i\rangle\exp[i(E_f-E_i)t]\right|^2, \quad \text{(B1)}$$

where E_f is the energy of the final state and E_i is the energy of the initial state. In the limit of an interaction time that is short compared to the typical energy of the atomic system, the energy phase in the matrix element can be approximated by unity. Then the transition probability simplifies to

$$P = \left| -i\int_{t_0}^{t_e} dt\langle\phi_f|\mathbf{r}\cdot\mathbf{E}(t)|\phi_i\rangle\right|^2 = |\langle\phi_f| - i\mathbf{q}\cdot\mathbf{r}|\phi_i\rangle|^2. \quad \text{(B2)}$$

In the lowest nonvanishing order of Magnus approximation in the length gauge, the first-order Magnus term, the transition probability is given by Eq. (21); and in the limit of weak pulses ($\mathbf{q}\cdot\mathbf{r} \ll 1$) gives an identical expression to Eq. (B2).

Second, let us consider a pulse with zero-momentum kick $\mathbf{q}=\mathbf{0}$ but with a nonvanishing displacement term $\boldsymbol{\alpha} \neq \mathbf{0}$. The first-order perturbation theory for short and weak pulses again yields for the transition probability Eq. (B1). It is now possible to perform a partial integration where the boundary term vanishes due to the zero-momentum kick condition. This yields

$$P = \left| -(E_f-E_i)\int_{t_0}^{t_e} dt\mathbf{A}(t)\cdot\langle\phi_f|\mathbf{r}|\phi_i\rangle\exp[i(E_f-E_i)t]\right|^2, \quad \text{(B3)}$$

with the matrix element identity $\langle\phi_f|\mathbf{p}|\phi_i\rangle = i(E_f - E_i)\langle\phi_f|\mathbf{r}|\phi_i\rangle$ valid for atomic eigenstates; the probability can be written as

$$P = \left| -i\int_{t_0}^{t_e} dt\langle\phi_f|\mathbf{p}\cdot\mathbf{A}(t)|\phi_i\rangle\exp[i(E_f-E_i)t]\right|^2. \quad \text{(B4)}$$

Now again assuming the interaction time to be short, so that the phase factor is close to unity, the transition probability can be approximated by

$$P = |\langle\phi_f| - i\mathbf{p}\cdot\boldsymbol{\alpha}|\phi_i\rangle|^2. \quad \text{(B5)}$$

The corresponding transition probability in the Magnus approximation considering only the leading (second-order) term in T is given by Eq. (24) and after linearization of the exponent, valid for weak pulses, agrees with the perturbation-theory result (B5).

Finally, we consider the action of laser pulses with $\mathbf{q}=\boldsymbol{\alpha}=\mathbf{0}$ but $\bar{\boldsymbol{\alpha}} \neq \mathbf{0}$ [type (iii) pulses considered in Sec. IV C]. Starting with Eq. (B4), performing partial integration of the transition amplitude, one obtains

$$P = \left| -(E_f-E_i)\int_{t_0}^{t_e} dt\boldsymbol{\alpha}(t)\cdot\langle\phi_f|\mathbf{p}|\phi_i\rangle\exp[i(E_f-E_i)t]\right|^2. \quad \text{(B6)}$$

Using the matrix identity $\langle\phi_f|\nabla V(\mathbf{r})|\phi_i\rangle = -i(E_f - E_i)\langle\phi_f|\mathbf{p}|\phi_i\rangle$ and assuming a short laser pulse yields for the transition probability,

$$P = |\langle\phi_f| - i(\nabla V)\cdot\bar{\boldsymbol{\alpha}}T|\phi_i\rangle|^2, \quad \text{(B7)}$$

which is just what one obtains from the linearization of the exponent in Eq. (48), valid in weak fields, in the third-order Magnus term.

A striking observation that can be made from the Eqs. (B2), (B5), and (B7) above is that the condition for breakdown of the first-order time-dependent perturbation depends on the type of the pulse. In the case with nonzero $\mathbf{q}$ the condition that Eq. (B2) is valid is $\mathbf{q}\cdot\mathbf{r} \ll 1$ which is most severe, i.e., the time-dependent perturbation theory breaks down for relatively small field amplitudes. For pulses with zero $\mathbf{q}$ and nonzero $\boldsymbol{\alpha}$ the condition $\mathbf{p}\cdot\boldsymbol{\alpha} \ll 1$ is valid, and it is less restrictive since $\boldsymbol{\alpha}$ is one order of pulse duration higher than $\mathbf{q}$. Finally, the least restrictive is $\nabla V\cdot\bar{\boldsymbol{\alpha}}T \ll 1$, which comes from Eq. (B7) and is valid for pulses for where both $\boldsymbol{\alpha}$ and $\mathbf{q}$ are zero.

In summary we have shown that for the three pulse types considered in Sec. IV the perturbation theory and the Magnus approximation yield identical transition probabilities in the limit of short and weak driving fields. In addition, we have shown that the validity of perturbation theory for short times depends strongly on the pulse properties.

2. Relation to the MMA

An approach very much related to the strategy developed in this paper is the modified Magnus approximation given in Ref. [2]. This approximation is based on the simple idea to subdivide the interaction time of the laser pulse into time slots corresponding to each half cycle of the pulse and then use the first Magnus approximation in the form of the operator Eq. (20) to give the TEO within each half cycle. The action of the total pulse is then given by the successive action of the sequence of half-cycle pulses. The TEO of the total pulse is then a product of half-cycle TEOs themselves approximated by the first-order Magnus term. This procedure was used in Ref. [2] to give the TEO,

$$U = \prod_{i=1}^{n} U_{\mathrm{hc},n-i+1}^{(1)}, \tag{B8}$$

where

$$U_{\mathrm{hc},i}^{(1)} = \exp[-iH_0\tau/2]\exp[-i\mathbf{q}_i\cdot\mathbf{r}]\exp[-iH_0\tau/2] \tag{B9}$$

are the half-cycle TEOs and τ is the half-cycle duration. The operator of Eq. (B8) is the *exact* TEO for a pulse consisting of δ functions in time, representing the momentum kicks delivered by each half cycle of the pulse. Since the full atomic propagator $\exp[-iH_0\tau]$ operates between half-cycle momentum kicks, the evaluation of the transition matrix element is difficult in general. Therefore further approximations were applied. First the atomic potential V was neglected in the propagation between δ-function kicks, which is justified for large laser field strength. Second, a linearization of the terms in the exponent of the TEO was carried out, which can be interpreted as a neglect of the spreading of the continuum electron wave packet. After these two steps the TEO is very simple, and the corresponding occupation probabilities of the initial state after each half cycle can be obtained in closed form for the hydrogenic states. The above procedure is called the asymptotic MMA, where "asymptotic" refers to the large laser field strength that is assumed for the validity of the approximations. Applying the above approximation it is possible to calculate accurate ionization and excitation probabilities even after many-cycle pulses [1,2]. Interestingly, the analytic formula agrees with numerical results even when the fields are not strong. This asymptotic procedure can be made also for pulses for which the total momentum transfer is zero [$\mathbf{q}=\mathbf{0}$, type (ii) pulses]. In this case the asymptotic MMA yields the transition probability

$$P_{\mathrm{MMA}} = \left| \int d^3\mathbf{r}\phi_f(\mathbf{r})\phi_i(\mathbf{r}+\Delta\mathbf{r}) \right|^2 = |\langle\phi_f|\exp[-i\mathbf{p}\cdot\Delta\mathbf{r}]|\phi_i\rangle|^2, \tag{B10}$$

where $\Delta\mathbf{r}$ is the displacement of a classical free electron with the field modeled as δ functions in time. This displacement is in general different to the displacement $\boldsymbol{\alpha}$ appearing in Eq. (24), which is the displacement of the classical free electron caused by the field $\mathbf{E}(t)$ *without* modeling the field by δ functions in time. So, in general for fields with envelope there is a difference between the MMA and the optimum-split third-order Magnus expansion result (24) formulated in this paper.

APPENDIX C: DERIVATION OF Eq. (36)

The splitting of the exponentials involving the atomic Hamiltonian H_0 in order to derive Eq. (36) from Eq. (9) is done with the help of the Zassenhaus formula [Eq. (6)].

In the case of an asymmetric half-cycle pulse the Zassenhaus formula has to be applied twice in Eq. (9). First with $A=-i\lambda H_0$ and $B=-i(1-\lambda)H_0-i\mathbf{q}\cdot\mathbf{r}/\tau-i\mathbf{p}\cdot(\mathbf{q}/2+\boldsymbol{\alpha}/\tau)+i(\boldsymbol{\nabla}V)\cdot(\boldsymbol{\alpha}/2+\mathbf{q}\tau/12-\overline{\boldsymbol{\alpha}})-i[q^2/12+\Phi/\tau+\boldsymbol{\alpha}\cdot\mathbf{q}/(2\tau)]$ which yields, for the TEO of the asymmetric half-cycle pulse ($t_e=t_0+\tau$),

$$\begin{aligned} U_{\mathrm{hc}}(t_e,t_0) = \exp[-i\lambda H_0\tau]\exp[&-i(1-\lambda)H_0\tau - i\mathbf{q}\cdot\mathbf{r} \\ &- i\mathbf{p}\cdot(\mathbf{q}\tau/2+\boldsymbol{\alpha}) + i(\boldsymbol{\nabla}V)\cdot(\boldsymbol{\alpha}/2+\mathbf{q}\tau/12-\overline{\boldsymbol{\alpha}})\tau \\ &- i(q^2\tau/12+\Phi+\boldsymbol{\alpha}\cdot\mathbf{q}/2)]\exp[-i\mathbf{p}\cdot\mathbf{q}\lambda\tau/2] \\ &\times\exp\{i(\boldsymbol{\nabla}V)\cdot\mathbf{q}\tau^2[\lambda^2/6+\lambda(1-\lambda)/3]+i\lambda q^2\tau/3\}. \end{aligned} \tag{C1}$$

The second application of Eq. (6) is with $A=-i\mathbf{q}\cdot\mathbf{r}/\tau-i\mathbf{p}\cdot(\mathbf{q}/2+\boldsymbol{\alpha}/\tau)+i(\boldsymbol{\nabla}V)\cdot(\boldsymbol{\alpha}/2+\mathbf{q}\tau/12-\overline{\boldsymbol{\alpha}})-i[q^2/12+\Phi/\tau+\boldsymbol{\alpha}\cdot\mathbf{q}/(2\tau)]$ and $B=-i(1-\lambda)H_0$ and yields

$$\begin{aligned} U_{\mathrm{hc}}(t_e,t_0) = \exp[&-i\lambda H_0\tau]\exp[-i\mathbf{q}\cdot\mathbf{r}-i\mathbf{p}\cdot(\mathbf{q}\tau/2+\boldsymbol{\alpha}) \\ &+ i(\boldsymbol{\nabla}V)\cdot(\boldsymbol{\alpha}/2+\mathbf{q}\tau/12-\overline{\boldsymbol{\alpha}})\tau - i(q^2\tau/12+\Phi \\ &+ \boldsymbol{\alpha}\cdot\mathbf{q}/2)]\exp[-i(1-\lambda)H_0\tau]\exp[i\mathbf{p}\cdot\mathbf{q}(1 \\ &- 2\lambda)\tau/2]\exp[i(\boldsymbol{\nabla}V)\cdot\mathbf{q}(-\lambda^2/2+\lambda-1/3)\tau^2 \\ &+ i(\lambda/2-1/6)q^2\tau]\exp[O(\tau^4)]. \end{aligned} \tag{C2}$$

When we further use the relation between two operators A and B,

$$\exp[A\tau]\exp[B\tau] = \exp[B\tau]\exp[A\tau]\exp[[A,B]\tau^2]\exp[O(\tau^3)], \tag{C3}$$

with $A=-i(1-\lambda)H_0$ and $B=i\mathbf{p}\cdot\mathbf{q}(1-2\lambda)$, Eq. (C2) can be rewritten as

$$\begin{aligned} U_{\mathrm{hc}}(t_e,t_0) = \exp[&-i\lambda H_0\tau]\exp[-i\mathbf{q}\cdot\mathbf{r}-i\mathbf{p}\cdot(\mathbf{q}\tau/2+\boldsymbol{\alpha}) \\ &+ i(\boldsymbol{\nabla}V)\cdot(\boldsymbol{\alpha}/2+\mathbf{q}\tau/12-\overline{\boldsymbol{\alpha}})\tau - i(q^2\tau/12+\Phi \\ &+ \boldsymbol{\alpha}\cdot\mathbf{q}/2)]\exp[i\mathbf{p}\cdot\mathbf{q}(1-2\lambda)\tau/2]\exp[-i(1 \\ &- \lambda)H_0\tau]\exp[i(\boldsymbol{\nabla}V)\cdot\mathbf{q}\tau^2(\lambda^2/2-\lambda/2+1/6) \\ &+ i(\lambda/2-1/6)q^2\tau]\exp[O(\tau^4)]. \end{aligned} \tag{C4}$$

With the definition of λ this expression can be further simplified and leads to Eq. (36).

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