Sudden perturbation of hydrogen atoms by intense ultrashort laser pulses

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We study theoretically how hydrogen atoms respond to intense ultrashort laser pulses of duration τ shorter than the inverse of the initial-state energy ε_i^{-1} . An analytical expression for the evolution operator \hat{S} is derived up to the first order of the sudden perturbation approximation. This approximation treats the laser-atom interaction beyond the dipole approximation and yields \hat{S} as a series in the small parameter $\varepsilon_i \tau$. It is shown that the effect of realistic laser pulses on atoms begins at the first order of $\varepsilon_i \tau$. Transitions between atomic (nlm) states of different *m* become possible due to the action of the pulse's magnetic field. Transitions between states of same *m* and arbitrary *l* become possible if the static Coulomb potential is taken into account during the pulse.

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I. INTRODUCTION

Hentschel *et al.* [1] and Paul *et al.* [2] have recently reported the generation of subfemtosecond (attosecond) light pulses. Subsequently it was demonstrated that in single attosecond pulses the wave form can be controlled $\lceil 3 \rceil$. This allows direct monitoring and manipulation of atomic electron processes in real time $[3-6]$, of interest in various fields of physics and technology such as nonlinear optics, coherent control of matter, and ultrashort high-power laser technology.

The response of atomic targets to ultrashort laser pulses has been considered theoretically in many papers $[7-14]$. The majority of these works rely on the dipole approximation which neglects the spatial-coordinate dependence of the laser field. This approximation is justified for not too intense pulses when the characteristic size of the laser wave variation is much larger than the characteristic size of the initially occupied atomic state. For example, this is the case for femtosecond pulses of intensities $10^{14} - 10^{15}$ W cm⁻² in the visible spectral range interacting with atoms in the ground state. These are the conditions typical in high-order harmonic generation experiments [15].

The dipole-approximation description of the laser-atom interaction needs to be corrected when the characteristic size of the laser-pulse variation is comparable with the atomic size. This is well known for atomic photoionizaton with lowintensity x-ray beams [16]. For nonintense ultrashort laser pulses the influence of the magnetic-field component on the electron dynamics can be neglected $[17,18]$. However, for very intense pulses the magnetic-field action should be taken into account [19–22]. This field induces significant electron drift along the propagation direction which can break down the atomic stabilization $[23]$ and significantly modify the induced photoemission spectra [12,24].

In this paper we study theoretically how a hydrogen atom interacts with an intense ultrashort pulse of duration $\tau < 1/\varepsilon_i = 2n_i^2$ where n_i is the principal quantum number of the initial state. For example, this condition is met if n_i >10 for available few-cycle femtosecond pulses of duration

 τ \sim 200 a.u. For available attosecond pulses (τ \sim 3 a.u.) it is satisfied for $n_i \geq 2$.

To describe the atomic response to such a pulse one has to take into account the spatial and time dependences of the electromagnetic field. This problem cannot be solved analytically. Under the considered conditions its numerical analysis is also exceptionally challenging, requiring considerable computer time and memory. To use numerical simulations one needs a good understanding of the laser-atom interaction physics. This is where approximate analytical theories can be indispensable. They are able to provide insight into the dynamics of the system and give benchmarks for more sophisticated numerical approaches. In this paper we employ the sudden perturbation approximation (SPA) [25] to get an expression for the evolution operator *S ˆ* for a hydrogen atom interacting with a strong ultrashort laser pulse. This approach allows consideration of the effects due to the finite-sizedpulse propagation through the quantum system and the Lorentz-force action with exact accounting of the field timespatial dependence. We use atomic units throughout unless specified otherwise.

II. THEORY

A. Problem statement

Consider a hydrogen atom subjected to an intense ultrashort electromagnetic pulse. Let the atomic electron be initially in state $|i\rangle$. To describe evolution of the electron wave function $|\psi\rangle$ we shall use the nonrelativistic timedependent Schrödinger equation

$$
i\frac{\partial}{\partial t}|\psi\rangle = [\hat{H}_0 + \hat{V}(\eta)]|\psi\rangle,\tag{1}
$$

where $\hat{H}_0 = -(1/2)\nabla^2 + V_{at}(r)$ is the unperturbed Hamiltonian, $V_{\text{at}}(r) = -1/|r| = -1/r$ is the Coulomb potential, and \hat{V} is the electron-field interaction potential,

$$
\hat{V}(\eta) = -A(\eta) \cdot \hat{p} + A^2(\eta)/2, \qquad (2)
$$

and where \hat{p} is the momentum operator, A is the vector po-*Electronic address: A.Lugovskoy@murdoch.edu.au tential of the field related to the electric field of the wave by

 $E = -\partial A / \partial t$, and $A = |A|$. For a traveling laser pulse the time and spatial coordinates appear in *A* in combination $\eta = t$ $-\alpha x$ where $\alpha = 1/c \approx 1/137$ and *c* is the speed of light. We assume that the *x* axis is directed along the pulse propagation. The electromagnetic field is assumed to be transversal with arbitrary polarization. Hence all capitalized vectors lie in the *yz* plane.

In the framework of the dipole approximation one is free to use different gauge-invariant descriptions of the laseratom interaction depending on which may be more useful for calculations. One should note that this is not possible here due to the explicit field dependence on the spatial coordinates. The other known gauge-invariant equations cannot be derived from Eq. (1) without additional approximations [26].

The electric field action on a free electron can be characterized by its displacement and drift momentum. They are, respectively,

$$
\Delta \boldsymbol{R} = \int_{-\pi/2}^{\pi/2} A(\eta) d\eta, \qquad (3)
$$

$$
\Delta P = -\int_{-\pi/2}^{\pi/2} E(\eta) d\eta = A(\pi/2). \tag{4}
$$

Also, due to the Lorentz force $f_L \approx (\alpha/2)dA^2(\eta)/d\eta$ [22,23], the electron additionally acquires displacement and momentum

$$
\Delta x \approx \frac{\alpha}{2} \int_{-\pi/2}^{\pi/2} A^2(\eta) d\eta,
$$
 (5)

$$
\Delta p_x \approx \int_{-\pi/2}^{\pi/2} f_L(\eta) d\eta = \frac{\alpha A^2(\pi/2)}{2},\tag{6}
$$

in the direction of the pulse propagation. In intense laser fields the electron-nuclear interaction can often be considered as a small perturbation. One can expect that the transformed electron wave function depends on the electromagnetic-field characteristics through these four parameters for very short pulses.

B. Restrictions on real laser pulses

Theoretically the pulse form can be chosen arbitrarily. There are many papers where atomic systems are subjected to fields producing nonzero ΔR or ΔP [13,27–30]. Recently, it has been realized that real laser pulses should satisfy the condition $\Delta R = \Delta P = 0$ [31]. Madsen [32] shows theoretically that $\Delta P=0$ for any real pulse which has to be localized spatially in three dimensions. Also, Muller [33] points out that, due to some experimental constraints imposed by the optical media generating the pulses, both drift momentum ΔP and electron displacement ΔR should be zero.

These are the restrictions we should impose on the form of theoretical ultrashort laser pulses to be realistic. Nevertheless, sometimes it is appropriate to consider pulses delivering nonzero ΔP and ΔR . For example, this is the case for halfcycle pulses interacting with Rydberg atoms [34]. Such a pulse consists of a very short main half-cycle followed by a long low-amplitude half-cycle pulse of opposite polarity. As one can expect, the integral (4) of both half-cycle parts is zero. However, under certain conditions [34], only the "kick" of the short half-cycle should be accounted for and the action of the long part of the full pulse can be neglected.

For this reason and in order to compare the SPA approach used with the results of other theoretical methods, we give derivations for the more general case of nonzero ΔR or ΔP . Then we come back to the case $\Delta R = \Delta P = 0$, thus concentrating on a hydrogen atom (de)excitation by realistic attosecond and femtosecond laser pulses. The Rydberg-atom interaction with half-cycle pulses will not be considered in this paper. It is currently under consideration and will be presented separately.

To study the dependences of the electron transition probabilities on the laser-pulse characteristics we assume, without loss of generality, that the electromagnetic field is

$$
E(\eta) = E_0 f(\eta) \begin{cases} \hat{y} \cos(\omega \eta + \varphi) \cos \xi + \hat{z} \sin(\omega \eta + \varphi) \sin \xi, & \text{if } |\eta| \le \pi/2, \\ 0, & \text{otherwise,} \end{cases}
$$
(7)

where E_0 is the field magnitude, $f(\eta)$ is the pulse envelope, ω is the carrier frequency, φ is the so-called carrier-envelope phase, and ξ determines the field polarization. For the vector potential *A* we write

$$
A(\eta) = -\int_{-\infty}^{\eta} E(\eta') d\eta', \qquad (8)
$$

which ensures that both *A* and *V* vanish in the area of atom localization in the infinite past. In the post-interaction region defined by the condition $\eta = t - \alpha x > \tau/2$ the residual values

of $A = A_1 = A(\tau/2)$ and $\hat{V} = \hat{V}_1 = \hat{V}(\tau/2)$ should also be zero for real laser pulses as follows from a previous discussion and Eqs. (2) and (4). This imposes an additional condition on the carrier frequency or carrier-envelope phase φ . It will be specified in the numerical-example section.

C. Sudden perturbation approximation

To find transition amplitudes from the initial state $|i\rangle$ to some final state \ket{f} we employ the SPA. This approximation yields the evolution operator \hat{S} corresponding to Eq. (1) as a

series over a small dimensionless parameter $\varepsilon \tau_{\text{int}}$ where τ_{int} $\approx \tau + 2n_i^2/c$ is the laser-atom interaction time [17] and ε correponds to a typical eigenenergy of Hamiltonian H_0 for the states involved in the processes of laser-atom interactions. These states include the initial state, the final state, and all intermediate states which can be excited during the pulse action with sufficiently large probability. This implies that the SPA validity depends on the result, as in the case of other perturbation theories. Here we assume that the laser pulse is sufficiently short so that

$$
\varepsilon_j \tau_{\text{int}} \ll 1 \tag{9}
$$

for any physically significant state $|j\rangle$. Another condition which should be met is imposed on the perturbation potential *V ˆ*. It is

$$
\left[\hat{V}(\eta), \hat{V}(\eta')\right] = \hat{V}(\eta)\hat{V}(\eta') - \hat{V}(\eta')\hat{V}(\eta) = 0 \tag{10}
$$

for any η and η' . One can see that this is the case for \hat{V} of Eq. (2) if the field is transversely polarized.

The general expression for the transition amplitudes from the initial state $|\phi_i\rangle$ to some final state $|\psi_f\rangle$ reads

$$
a_{fi} = \langle \psi_f | \hat{S}_f(\infty, t_0) e^{iH_f t_0} e^{-iH_i t_0} \hat{S}_i(t_0, -\infty) | \phi_i \rangle, \qquad (11)
$$

where $\hat{H}_i = \hat{H}_0$, $\hat{H}_f = \hat{H}_0 + \hat{V}_1$, $|\phi_i\rangle$ ($|\psi_f\rangle$) is an eigenfunction of $\hat{H}_{i(f)}$, and t_0 corresponds to the center of the time interval τ_{int} outside which the field action on the atom disappears. Equation (11) takes into account the fact that the finalchannel Hamiltonian is different from the initial-channel Hamiltonian due to the presence of the residual dc field *A* $=A_1$ in the region occupied by the atom. The evolution operator $\hat{S}_{i(f)}(t, t')$ can be estimated in the zeroth order in $\epsilon \tau$ by

$$
\hat{S}_{i(f)}^{(0)}(t,t') = \exp[i\hat{H}_{i(f)}t_0] \exp\left[-i\int_{t'}^{t}\hat{V}_{i(f)}(t)dt\right]
$$

$$
\times \exp[-i\hat{H}_{i(f)}t_0],
$$
\n(12)

where $\hat{V}_i = \hat{V}(\eta)$ and $\hat{V}_f = \hat{V}(\eta) - \hat{V}_1$. Also, in what follows, we shall use the first-order expression for the \hat{S} operators,

$$
\hat{S}_{i}^{(1)}(t,t') = \hat{S}_{i}^{(0)}(t,t') [I + i\hat{\Delta}_{i}(t,t')] ,
$$

$$
\hat{S}_{f}^{(1)}(t,t') = [I + i\hat{\Delta}_{f}(t,t')] \hat{S}_{f}^{(0)}(t,t') ,
$$
 (13)

where

$$
\hat{\Delta}_i(t,t') = -\int_{t'}^{t} [\hat{S}_i^{(0)}(\tau,t')]^{-1} \hat{W}_i(\tau) \hat{S}_i^{(0)}(\tau,t') d\tau, \qquad (14)
$$

$$
\hat{\Delta}_f(t,t') = -\int_{t'}^{t} \left[\hat{S}_f^{(0)}(t,\tau) \right]^{-1} \hat{W}_f(\tau) \hat{S}_f^{(0)}(t,\tau) d\tau, \qquad (15)
$$

and

$$
\hat{W}_{i(f)}(t) = it[\hat{H}_0, \hat{V}_{i(f)}(t - \alpha x)].
$$
\n(16)

With the use of expression (2) and taking into account that $\ket{\psi_f} = \exp[iA_1 \cdot r] \ket{\phi_f}$ one gets, from Eqs. (11) and (12),

$$
a_{fi}^{(0)} = \langle \phi_f | e^{i\Delta p \cdot r} \exp[i\Delta R_1(x) \cdot \hat{p} - i\gamma_1] | \phi_i \rangle, \qquad (17)
$$

where $\Delta p = \hat{x} \Delta p_x - \Delta P$, $\Delta R_1(x) = \Delta R - [\tau/2 + \alpha x]A_1$, $\gamma_1 = \gamma$ $-1A_1^2 \tau/4$, and $\gamma = \int_{-\pi/2}^{\pi/2} A^2(\eta)/2d\eta$. Equation (17) shows that, in the zeroth approximation, the pulse action on the atomic electron would result in acquiring a drift momentum Δp due to the combined action of the electric- and magnetic-field components. It also includes electron displacement by ΔR_1 dependent on *x* and wave-function rotation in the complex plane determined by the phase γ_1 . We see that, in the general case, corrections due to the action of the wave magnetic field appear already in the zeroth approximation.

If both $\Delta \mathbf{R}$ and $\Delta \mathbf{P}$ are zero, then Eq. (17) reduces to

$$
a_{fi}^{(0)} = \delta_{i,f} e^{-i\gamma},\tag{18}
$$

where $\delta_{i,f}$ is the Kronecker delta (note that $\Delta p_x = \alpha A_1^2 / 2 = 0$). We see that, in the zeroth order, physical pulses only change the wave-function phase by a constant value γ and, hence, no transitions between different states occur irrespective of the field magnitude. To calculate the probability of such transitions one needs higher orders of the SPA. The following is an expression for the first-order transition amplitude $a_{fi}^{(1)}$ applicable for any A_1 :

$$
a_{fi}^{(1)} = \langle \phi_f | e^{-i\Delta p_z z} (\hat{I} + i\hat{\Delta}_f) \hat{S}_f^{(0)}(\infty, 0) \hat{S}_i^{(0)}(0, -\infty) (\hat{I} + i\hat{\Delta}_i) | \phi_i \rangle, \tag{19}
$$

where \hat{I} is the identity operator,

$$
\hat{\Delta}_{i(f)} = \int_{t'_{i(f)}}^{t_{i(f)}} dt t \frac{\partial}{\partial t} \left[V_{\text{at}}(|\mathbf{r} - \mathbf{R}_{i(f)}(\boldsymbol{\eta})|) + \alpha \hat{V}_{i(f)}(\boldsymbol{\eta}) \hat{p}_x + \frac{\alpha^2 \hat{V}_{i(f)}(\boldsymbol{\eta})}{2} + i \frac{\alpha^2 \hat{V}_{i(f)}(\boldsymbol{\eta})}{2} \right],
$$
\n(20)

and where $t'_{i(f)} = -\infty(0)$, $t_{i(f)} = 0(\infty)$,

$$
\boldsymbol{R}_i(\boldsymbol{\eta}) = \int_{-\infty}^{\eta} A(\boldsymbol{\eta}) d\boldsymbol{\eta}, \qquad (21)
$$

$$
\boldsymbol{R}_f = \int_{-\infty}^{\eta} \left[A(\eta) - A_1 \right] d\eta. \tag{22}
$$

 $a_{fi}^{(1)} = [\delta_{i,f} + i \langle \phi_f | \hat{\Delta}_0 | \phi_i \rangle] e^{-i \gamma}$ (23)

 (23)

For physical pulses $(\Delta R = 0 \text{ and } \Delta P = 0)$ one has, from Eqs. (19) and (20) ,

and

$$
\hat{\Delta}_0 = -\Delta x \hat{p}_x - \frac{\alpha^2}{2} \int_{-\pi/2}^{\pi/2} V^2(\eta) d\eta
$$

+
$$
\int_{-\pi/2}^{\pi/2} (\eta + \alpha x) A(\eta) \cdot \nabla V_{\text{at}}(r - R_i(\eta)) d\eta. \quad (24)
$$

The first term on the right-hand side of Eq. (24) appears due to the action of the field magnetic component on the electron driven by the electric field of the wave. Physically it is related to the electron displacement in the pulse-propagation direction. This term is of the order of α ($\Delta x = \alpha \Delta \phi$). It allows electron transitions between states with different magnetic quantum numbers.

The second term on the right-hand side (RHS) of Eq. (24) is of the second order in α . It characterizes the influence of the pulse magnetic field on the electron motion induced by the magnetic field itself. If the field is sufficiently low and satisfies the condition $\alpha A_0 \ll 1$, the second term can be neglected. In this limit one can characterize the magnetic-field action with parameter Δx only. Moreover, this condition is necessary for convergence of the SPA since, as one can show, higher orders of this term appear in the next orders of the SPA. In this paper we will assume that the condition αA_0 ≤ 1 is met.

The third term of Eq. (24) takes into account the Coulomb potential variation during the pulse which is neglected in the zeroth order of the SPA. This can be seen from the equality

$$
\int_{-\pi/2}^{\pi/2} (\eta + \alpha x) A(\eta) \cdot \nabla V_{\text{at}}(\mathbf{r} - \mathbf{R}_i(\eta)) d\eta
$$

$$
= \int_{-\pi/2}^{\pi/2} [V_{\text{at}}(\mathbf{r}) - V_{\text{at}}(\mathbf{r} - \mathbf{R}_i(\eta))] d\eta. \tag{25}
$$

In what follows we will refer to the third term of Eq. (26) as a Coulomb-force correction. This is the only term in $\hat{\Delta}_0$ which depends on the position r of the electron. As far as it is a function of the electron trajectory \mathbf{R}_i one can expect a polarization dependence for the induced transition probabilities. Note that the third term on the RHS of Eq. (24) is not necessarily dependent on α since the SPA expansion parameter is $\varepsilon \tau$.

III. RESULTS

In this section we study numerically the effect of the ultrashort electromagnetic pulse on a hydrogen atom and consider the example of bound-bound transitions.

A. Laser pulse

We assume that the electromagnetic field satisfies the conditions for real physical pulses and, hence, both ΔP and ΔR are zero. Let the pulse be linearly polarized and its vector potential *A* be directed along the *z* axis. Without loss of generality we use the expression

$$
A(\eta) = \hat{z}A_0 \sin(\omega \eta + \varphi) \begin{cases} \cos^2(\pi \eta/\tau), & \text{if } |\eta| \le \pi/2, \\ 0, & \text{otherwise.} \end{cases}
$$
 (26)

This form is often used in the literature for few-cycle femtosecond pulses $[7,13,35,36]$.

Expression (26) ensures that $E(\pm \tau/2)$ and $A(\pm \tau/2)$ are equal to zero and, hence, $\Delta P = A(\tau/2) = 0$ for any laser pulse parameters A_0 , τ , ω , and φ . To satisfy the second condition $(\Delta R = 0)$ we choose the laser frequency ω to be ω $=2\pi n_{\rm osc}/\tau$ where $n_{\rm osc}$ is a positive integer corresponding to the number of field oscillations during the pulse. Also the condition $\Delta R = 0$ requires that $n_{\text{osc}} \geq 2$ for a field given by Eq. (26). The vector potential magnitude A_0 is related to the magnitude of the pulse's electric-field component as A_0 $=E_0 / \omega = E_0 \tau / (2 \pi n_{\text{osc}})$. Taking into account that $\alpha A_0 \le 1$ we see that E_0 and τ have to satisfy additionally the condition $E_0 \tau \ll 2\pi n_{\text{osc}}c$. Thus, for the electromagnetic pulse (26), we have four independent governing parameters: the electricfield magnitude E_0 , pulse duration τ , field-oscillation number n_{osc} , and carrier envelope phase φ .

B. Nonmagnetic transitions

To estimate the magnitude of the ultrashort laser pulse effect on the nonmagnetic transitions we consider the case of a linearly polarized laser pulse given by Eq. (26). We assume that the pulse comprizes only two oscillations $(n_{osc}=2)$ and the field magnitude can be higher than the atomic field. Presently, these conditions can be met for femtosecond laser pulses. For these pulses the SPA is applicable for Rydberg atoms. Our calculations show that femtosecond pulses with Rydberg atoms have qualitatively similar behavior as attosecond pulses with atoms in low-excited states. For numerical demonstration, without loss of generality, we consider a femtosecond pulse on a hydrogen atom initially in a highly excited state.

The transition amplitudes given by Eq. (23) have a structure characteristic of the perturbation theory $[37]$. It can be applied to calculate the estimates for transition probabilities for different initial and final states. Also, for the SPA to converge, it is required that the effect of the correction term Δ_0 on the electron wave function has to be small. That is, we require $|\langle f | \Delta_0 | i \rangle| \ll 1$ for the given initial state $|i\rangle$ and any final state \ket{f} . This results in the additional restriction on the field magnitude E_0 .

The probabilities of transitions between states with the same magnetic quantum numbers (nonmagnetic transitions) can be estimated with the use of Eqs. (23) and (24) as follows:

$$
P_{i\to f}^{\text{nm}} = \left| \int_{-\pi/2}^{\pi/2} d\eta \eta A(\eta) \langle f | \partial_z V_{\text{at}}(\mathbf{r} - \mathbf{R}_i(\eta)) | i \rangle \right|^2 \qquad (27)
$$

or, alternatively,

$$
P_{i\to f}^{\text{nm}} = \left| \int_{-\pi/2}^{\pi/2} d\eta \langle f | V_{\text{at}}(\mathbf{r}) - V_{\text{at}}(\mathbf{r} - \mathbf{R}_i(\eta)) | i \rangle \right|^2, \quad (28)
$$

where $\partial_z = \partial/\partial z$. Particularly, for a transition between different s states, Eq. (27) can be simplified to the form

$$
P_{n_{i}s \to n_{f}s}^{\text{nm}} = \left| \int_{-\pi/2}^{\pi/2} d\eta \eta A(\eta) \frac{\mathcal{Z}_{i}(\eta)}{|\mathcal{Z}_{i}(\eta)|^{3}} \times \int_{0}^{|\mathcal{Z}_{i}(\eta)|} R_{n_{i},0}(r) R_{n_{f},0}(r) r^{2} dr \right|^{2}, \qquad (29)
$$

where n_f is the principal quantum number of the final state with the radial function $R_{n_f,0}$ and $\mathcal{Z}_i(\eta)$ is the *z* component of displacement $\mathbf{R}_i(\eta)$ $[\mathbf{R}_i(\eta) = \hat{z} \mathcal{Z}_i(\eta)]$. Leaving the first nonva-

FIG. 1. The probabilities of electron transitions between states with the same magnetic quantum numbers $(m_i = m_f = 0)$ as functions of the electric-field magnitude E_0 . The electromagnetic field used in calculations is given by Eq. (26) with pulse duration $\tau=1$ fs, field oscillation number $n=2$, and carrier-envelope phase $\varphi=0$. The initial state is 8*s*, and the final states are shown in the legend. Thick and thin lines correspond to probabilities calculated by Eq. (29) and its approximation (31), respectively.

nishing Taylor-expansion term of the integrand in Eq. (29), one can see that $P_{n_i s \to n_f s}^{\text{nm}}$ behaves, at small \mathcal{Z}_i , as follows:

$$
P_{n_i s \to n_f s}^{\text{nm}} \approx \frac{4}{9 n_i^3 n_f^3} \left(\int_{-\pi/2}^{\pi/2} \mathcal{Z}_i^2(\eta) d\eta \right)^2 \tag{30}
$$

$$
= \frac{\left[3n_{\text{osc}}^2(n_{\text{osc}}^2 - 1) + 4 + 2\cos(2\varphi)\right]^2}{2^{14}3^2\pi^8 n_{\text{osc}}^8(n_{\text{osc}}^2 - 1)^4} \frac{E_0^4 \tau^{10}}{n_i^3 n_f^3}.
$$
 (31)

where expression (31) was obtained for field (26). Approximation (30) is valid for $\mathcal{Z}_i \le 1$ as one can show by comparing the first and second Taylor-expansion terms of the integrand in Eq. (29). Equation (31) shows that, for fixed laser pulse parameters E_0 , τ , and φ , the probability $P_{n_i s \to n_f s}^{\text{nm}}$ decreases as $n_{\rm osc}^{-8}$ with an increase in the oscillation number $n_{\rm osc}$. Also, it has a very weak dependence on the carrier-envelope phase φ .

Expression (30) reveals the universal dependence of the *s*-*s* transition probabilities on the laser pulse parameters for small \mathcal{Z}_i , irrespective of the principal quantum numbers of the initial n_i and final n_f states. Specifically, for sufficiently small \mathcal{Z}_i probability, $P_{n_i s \to n_i s}^{\text{min}}$ behaves as $E_0^4 \tau^{10}$. We also see that these probabilities decrease rapidly with an increase in n_i and n_f . It means that the mechanism responsible for these bound-bound transitions results in preferable atom deexcitation. This is physically meaningful since the *S ˆ*-matrix correction due to the electron-nuclear interaction is peaked at the nuclear position as one can see from Eq. (24). So the closer the electron can approach the nucleus, the more preferable is such a transition.

These regularities are illustrated in Fig. 1. It shows the probabilities of $8s-n_f s$ transitions for different quantum numbers n_f of the final state as a function of the electric-field magnitude E_0 at $\tau = 1$ fs. For the other final states the transition probabilities shown as thick lines were calculated with the use of Eq. (29). The thin lines also shown in the figure were computed by Eq. (31) .

FIG. 2. The probabilities of nonmagnetic transitions 8*s* $\rightarrow 3l_f m_f = 0$ for carrier-envelope phases $\varphi = 0$ (thick lines) and φ $=\pi/2$ (thin lines), as functions of the electric-field magnitude E_0 . The electromagnetic-field parameters are $\tau=1$ fs and the field oscillation number $n_{\text{osc}}=2$.

We see, from Fig. 1, that transitions to the states with lower n_f are more probable for all values of E_0 . All the curves rise as E_0^4 with increasing E_0 while $E_0 \lessapprox 10^{-1}$. For larger field magnitudes the maximum electron displacement during the pulse max $[\mathcal{Z}_i]$ is comparable with or higher than 1 and we see deviations of all probabilities calculated by Eq. (29) from the power law (31). Note that $\max[\mathcal{Z}_i] \approx 10E_0$ in the considered case.

For transitions from the n_i s state to some state with l_f >0 one can derive

$$
P_{n_i s \to (n_f, l_f, 0)}^{\text{nm}} = \left\{ \int_{-\pi/2}^{\pi/2} d\eta \eta A(\eta) \frac{1}{\sqrt{l_f + 1}} \times \left[\int_{0}^{|\mathcal{Z}_i(\eta)|} dr R_{n_i, 0}(r) R_{n_f, l_f}(r) \times \left(\frac{(l_f + 1) r^{l_f + 2}}{|\mathcal{Z}_i(\eta)| [\mathcal{Z}_i(\eta)]^{l_f + 1}} + \frac{l_f [\mathcal{Z}_i(\eta)]^{l_f - 1}}{r^{l_f - 1}} \right) \right] \times \left[\int_{0}^{\infty} dr R_{n_i, 0}(r) R_{n_f, l_f}(r) \frac{l_f [\mathcal{Z}_i(\eta)]^{l_f - 1}}{r^{l_f - 1}} \right] \right\}^2.
$$
\n(32)

For small \mathcal{Z}_i these probabilities are determined by the second term of Eq. (32) and can be approximated by

$$
P_{(n_i,0,0)\to(n_f, l_f,0)}^{\text{nm}} \approx \frac{a_{n_i, n_f, l_f}}{l_f^2(l_f+1)} \left[\int_{-\pi/2}^{\pi/2} [\mathcal{Z}_i(\eta)]^{l_f} d\eta \right]^2, \quad (33)
$$

where

$$
a_{n_i, n_f} l_f = \left(\int_0^\infty R_{n_i, 0}(r) R_{n_f, l_f}(r) r^{1-l_f} dr \right)^2.
$$
 (34)

The nonmagnetic transition probabilities $P_{i\rightarrow f}^{\text{nm}}$ versus the electric-field magnitude E_0 for different final states $(n_f, l_f, 0)$ are shown in Fig. 2. We see that the probabilities $P_{8s\rightarrow 3s}^{\text{min}}$, $P_{8s\rightarrow 3p}^{\text{nm}}$, and $P_{8s\rightarrow 3d}^{\text{nm}}$ demonstrate a power-law dependence versus the electric-field strength E_0 at small \mathcal{Z}_i . Their slopes are, respectively, 4, 2, and 4. This is in agreement with the

FIG. 3. The specified values $|\langle f|\hat{p}_{x}|i\rangle|^{2}$ versus principal quantum number *n*. The dotted line corresponds to the function $f(n)$ $=0.0176n^{-2}$.

estimation $P_{(n_i,0,0)\to(n_f,l_f,0)}^{\text{nm}} \sim E_0^{2l_f} \tau^{2(2l_f+1)}$ which follows from Eq. (33). Due to this power law, transitions to the $3p$ state are more probable than the other transitions at small E_0 . When *E*₀ is sufficiently large $(E_0 > 2 \times 10^{-2})$, max $[\mathcal{Z}_i]$ \gtrsim 1, and Eqs. (30) and (33) are inapplicable, we see that $P_{8s\rightarrow 3s}^{\text{nm}}$ is almost one order of magnitude higher than the transition probabilities to the other states of the $n_f = 3$ shell. This is also generally the case for transitions to the 3*p* state in comparison with transitions to the 3*d* state. From the physical point of view this can be explained by the fact that the electron density of the states with nonzero orbital momentum *l* is reduced due to the factor r^{2l} near the nucleus where the major contribution to the probabilities comes from.

Figure 2 shows also the effect of varying carrier-envelope phase φ . The atomic response to few-cycle laser pulses depends on this parameter, which is of current interest in view of possible applications $[3,15,38,39]$. We have calculated many choices and present the two which represent the maximum variation. In the case of $P_{8s\rightarrow 3s}^{nm}$ the thick and thin lines are almost indistinguishable, indicating almost no φ dependence. Due to the symmetric properties of matrix element $\langle f|V(r) - V(r + R_i)|i\rangle$ and electromagnetic field *A*(η) of Eq. (26), the probability of transition between the state with odd $\Delta l = l_f - l_i$ is equal to zero for any E_0 at $\varphi = \pi/2$. For this reason the probability of $P_{8s\to 3p}^{nm}$ for $\varphi = \pi/2$ is exactly zero and hence not plotted. The $P_{8s\rightarrow 3d}^{\text{min}}$ does show some minor variation with φ .

C. Magnetic transitions

In addition to nonmagnetic transitions considered in the previous subsection an ultrashort pulse can induce transitions between states with different magnetic quantum numbers (magnetic transitions). In the first order of the SPA their probabilities read

$$
P_{i \to f}^{\mathbf{m}} = \Delta x^2 |\langle f | \hat{p}_x | i \rangle|^2. \tag{35}
$$

We see that $P_{i\rightarrow f}^{\text{m}}$ is a product of two factors. One of them $(|\langle f|\hat{p}_x|i\rangle|^2)$ specifies how the probabilities depend on the initial and final states. It determines the selection rules. Namely, $P_{i\rightarrow f}^{\text{m}}$ can be nonzero only for $\Delta l = l_f - l_i = \pm 1$ and $\Delta m = m_f$ $-m_i = \pm 1$.

Figure 3 presents $|\langle f|\hat{p}_{x}|i\rangle|^{2}$ calculated for different initial *s*

and final p_1 states (the subscript "1" indicates that $m_f = 1$). The curve with triangles shows $|\langle np_1| \hat{p}_x | i \rangle|^2$ calculated for the atom being initially in the 20*s* state. One can see that $|\langle np_1| \hat{p}_x | 20s \rangle|^2$ is strongly peaked around *n*=20. One should also note that $\langle 20p_1 | \hat{p}_x | 20s \rangle = 0$. More generally, our calculations indicate that $\langle f | \hat{p}_x | i \rangle = 0$ if the principal quantum numbers of the initial and final states are equal to each other. This means that intrashell magnetic transitions are prohibited in the first order of the SPA.

In Fig. 3 the $\left| \frac{\langle (n-1)p_1 | \hat{p}_x | ns \rangle}{\}^2$ and $\left| \frac{\langle (n+1)p_1 | \hat{p}_x | ns \rangle}{\}^2$ curves characterize magnetic transitions between states of the closest shells. We see that both decrease monotonically with an *n*−2 dependence. The probability of a transition to the upper shell is slightly bigger than the probability of a transition to the lower shell for $n < 20$. For $n > 20$ the difference is very small.

The other factor determining the transition probability (35) is Δx^2 . It accumulates the probability dependence of the laser pulse parameters. For the field of Eq. (26) one has Δx $= \alpha E_0^2 \tau^3 / (128 \pi^2 n_{\text{osc}}^2)$. Hence, the transition probability (35) is independent of the carrier-envelope phase φ , increases as I^2 with the laser intensity $I = E^2$, and has a strong dependence on the pulse duration τ .

D. SPA in relation to the other methods

Another approximate analytical approach which is used for the description of atom interactions with ultrashort laser pulses is the impulse approximation (IA) [40,41]. It is based on a series of gauge transformations of the Schrödinger equation, which allows several different approximations for the electron wave function. Particularly, in our notation, one of these forms reads $[40]$

$$
|\psi(t)\rangle_{IA} = e^{-i\hat{H}_0 t} e^{-i\Delta P(t)r} e^{i\delta(r,t)} e^{i\mathbf{R}'_i(t)\hat{p}} |\phi_i\rangle, \tag{36}
$$

where

$$
\mathbf{R}'_i(t) = \int_{-\pi/2}^t t' \mathbf{E}(t') dt' = \mathbf{R}_i(t) - t \Delta \mathbf{P}(t)
$$
(37)

is now the electron displacement during the pulse action and

$$
\delta(\mathbf{r},t) = \nabla V_{\text{at}}(\mathbf{r}) \cdot \int_{-\pi/2}^{t} (t'^2/2) E_i(t') dt' \tag{38}
$$

appears due to the Coulomb-force correction. Note also that the wave function of Eq. (36) was obtained in the framework of the dipole approximation and, hence, does not account for the magnetic-field effect.

If condition (9) is fulfilled for all essential states involved, one can drop the exponential factor with \hat{H}_0 in Eq. (36). Also assuming that the Coulomb-force correction is negligible one can see that expression (36) leads to the same transition amplitudes as the zero-order SPA in the dipole-approximation limit $(\alpha=0)$. Thus Eq. (17) generalizes the results of [42] for ultrashort pulses by taking into account the effect of the magnetic-field component.

As mentioned earlier, the physical laser pulses have no effect on the atom in the zeroth order of the SPA. This is where corrections due to the magnetic force and Coulomb force can be distinguishable. The SPA and IA give different expressions for the Coulomb-force correction. This difference comes from the approximation used in the IA $[41]$:

$$
\Delta V_{\text{at}}(\boldsymbol{r}, \boldsymbol{R}) = V_{\text{at}}(\boldsymbol{r}) - V_{\text{at}}(\boldsymbol{r} - \boldsymbol{R}) \approx \boldsymbol{R} \cdot \boldsymbol{\nabla} V_{\text{at}}(\boldsymbol{r}). \qquad (39)
$$

In this connection one should note that the transition amplitudes a_{fi} depend on the integral of ΔV_{at} [see Eqs. (24) and (25)] and, hence, the Taylor series expansion of ΔV_{at} should be valid for the whole space. This is not the case for the singular Coulomb potential V_{at} . For this reason the approximation (39) as well as the whole Taylor series for ΔV_{at} results in an incorrect expression for transition amplitudes between *s* states. Nevertheless, as our numerical calculations show, the IA gives a correct result for transitions between states with $|\Delta l|=1$. This is due to the fact that the electron density for at least one of the radial functions is zero at *r* =0, thus eliminating the contribution of the singularity area.

In the numerical example given in the previous section the most probable transition at sufficiently large E_0 is the $8s \rightarrow 1s$ transition. One should note that the applicability condition (9) is violated for the final 1*s* state $(\varepsilon_{1s}\tau \approx 21)$ $>$ 1). Nevertheless, the SPA can still be used for the transitions with $\varepsilon_f \tau \leq 1$ provided that the probability of an atom to remain in the initial state is much larger than the probabilities of any other transitions. This leads us back to the condition $|\langle f | \hat{\Delta}_0 | i \rangle| \ll 1$ required for the SPA to be convergent.

To show that this is the case one can transform the Schrödinger equation (1) into the form

$$
i\frac{\partial|\psi'\rangle}{\partial t} = -\frac{1}{2}\nabla^2|\psi'\rangle + \hat{V}_{\text{at}}(\mathbf{r})|\psi'\rangle + \hat{U}(\mathbf{r}, \eta)|\psi'\rangle, \qquad (40)
$$

where

$$
\hat{U}(r,\eta) = \hat{V}_{\text{at}}(r - R_i(\eta)) - \hat{V}_{\text{at}}(r) + \alpha \hat{V}(\eta)\hat{p}_x
$$

$$
-i\frac{\alpha^2}{2}\frac{\partial \hat{V}(\eta)}{\partial \eta} - \frac{\alpha^2}{2}[\hat{V}(\eta)]^2
$$
(41)

and

$$
|\psi'\rangle = \exp\left[i\int_{-\infty}^{\eta} \hat{V}(\eta')d\eta'\right]|\psi\rangle.
$$
 (42)

One can recognize in Eq. (42) the Kramers-Henneberger gauge transformation but generalized for the case of a spatially dependent field ($\eta = t - \alpha x$). This dependence gives rise to the α -dependent terms in Eq. (41) which we already see in Eq. (20) .

In the perturbation limit the terms in Eq. (41) of the α^2 order can be neglected. Then, for physical laser pulses, the transition probability between different *s* states due to the perturbation \hat{U} can be estimated as follows:

FIG. 4. Same as Fig. 1, but thin and thick lines now correspond to probabilities calculated by Eqs. (29) and (43), respectively.

$$
P_{i\to f}^{\text{nm}} = \left| \int_{-\infty}^{\infty} dt \exp[i\omega_{fi}t] \langle f|V_{\text{at}}(\mathbf{r}) - V_{\text{at}}(\mathbf{r} - \mathbf{R}_i(\boldsymbol{\eta}))|i\rangle \right|^2.
$$
\n(43)

One can see that this expression transforms to Eq. (28) for small transition frequencies $\omega_{fi} = \varepsilon_f - \varepsilon_i \ (\omega_{fi} \tau \ll 1).$

Figure 4 shows the probabilities of $8s-n_fs$ transitions for different quantum numbers n_f of the final state as functions of the electric-field magnitude E_0 at $\tau = 1$ fs. We see that the probabilities calculated by Eq. (43) deviate significantly from the SPA probabilities of Eq. (28) for transitions to the 1*s* ($\varepsilon_{1s}\tau \approx 21$) and 2*s* ($\varepsilon_{2s}\tau \approx 5$) states. The former probabilities are substantially smaller than the SPA probabilities. This is due to the highly oscillating factor in Eq. (43). We also see that for higher states with $\varepsilon_f \tau \lesssim 1$ both formulas are in good agreement.

Thus, for physical laser pulses $(\Delta P = \Delta R = 0)$ both approaches—the first-order SPA and the perturbation description in the Kramers-Henneberger frame—give similar analytical expressions for the transition amplitudes for the states satisfying condition (9). If laser pulses produce nonzero drift momentum ΔP , the SPA also yields compact analytical expressions for transitions amplitudes. However, the use of the perturbation approach in the Kramers-Henneberger frame can be much more complicated. This is due to the factor $exp[iA_1 \cdot r]$ in the wave functions of the final channel.

E. Discussion

In this subsection we first compare the results for magnetic and nonmagnetic transitions and then discuss possible applications of the SPA.

Previously we saw that deexcitation is the preferable process for nonmagnetic bound-bound transitions. This is in contrast with magnetic transitions which are dominated by transitions between states with $n_f - n_i = \pm 1$. This different behavior is due to the fact that these processes are initiated by sufficiently different mechanisms as demonstrated qualitatively in Fig. 5. Each figure shows three circles representing three states with principal quantum numbers n_{i-1} , n_i , and n_{i+1} . The circle radii are in a relation determined by the expression $r_n \approx 2n^2$ for the circle radius.

FIG. 5. (a) The diagram for an electron interaction with the magnetic-field pulse. The vectors show how the magnetic field displaces the electronic cloud denoted by the circle $|n_i s\rangle$. The vector length corresponds to displacement Δx . (b) Similar to (a) except that the vectors correspond to the additional electron momenta arising due to the accounting of the Coulomb force variation (see text).

Figure $5(a)$ shows the uniform displacement of the electron cloud by Δx in the *x* direction due to the Lorentz force. The vector length is equal to Δx . The displaced electron can stay in its initial state $|i\rangle$ or can be captured to some state $|f\rangle$ with $|l_f|=1$ and $|m_f|=1$. The amplitude of such a transition is of the order of $\langle \varphi_f(r) | \varphi_i(r + \hat{x} \Delta x) \rangle$. For small Δx this overlapping integral decreases with an increase in $|n_f - n_i|$ due to the wave-function spatial properties.

In the framework of classical physics one can relate the nonmagnetic transitions to the Coulomb force variation

$$
\Delta F_{\rm C} = F_{\rm C}(r(t)) - F_{\rm C}(r_{\rm st}(t)) = -\nabla [V_{\rm at}(r(t)) - V_{\rm at}(r_{\rm st}(t))],\tag{44}
$$

where $\mathbf{r}(t) = \mathbf{r}_{\text{st}}(t) + \mathbf{R}_i(t)$ is the true trajectory of the electron. This variation results in an additional electron momentum

$$
\delta p = \int_{-\pi/2}^{\pi/2} \Delta F_C(r(t))dt \approx \frac{\partial F_C(r(t))}{\partial z} \int_{-\pi/2}^{\pi/2} \mathcal{Z}_i(\eta) d\eta.
$$
\n(45)

Figure 5(b) shows the set of vectors $\partial F_C(r)/\partial z$ calculated for different points of the n_i s circle.

We see, from Fig. $5(b)$, that the vectors of the lower semicircle are focused towards the atomic nucleus. Taking into account the problem symmetry, one can see that the electron gains an additional momentum δp toward the nucleus if z 0 . Then it can be captured by one of the lower-excited states. If the electron is initially above the plane $z=0$, it is kicked outside. We suppose that this region contributes most to atomic ionization.

For small displacements \mathcal{Z}_i the relative contribution of both mechanisms can be compared with the use of Eqs. (31) and (35) and the estimate $|\langle f | \hat{p}_x | i \rangle|^2 \approx 0.0176 n_i^{-2}$ (see caption to Fig. 3). In this case,

$$
\frac{P_{n_i s \to (n_i \pm 1)p_1}^{\text{m}}}{P_{n_i s \to 1s}^{\text{nm}}} \approx \frac{10^{-3} n_{\text{osc}}^4 (n_{\text{osc}}^2 - 1)^4}{\left[3n_{\text{osc}}^2 (n_{\text{osc}}^2 - 1) + 4 + 2 \cos(2\varphi)\right]^2} \frac{n_i}{\tau^4}.
$$
\n(46)

For few-cycle laser pulses this expression predicts that the magnetic transitions between states with the principal quantum number $n_f = n_i \pm 1$ are far less effective than atom deexcitation from n_i s to the ground state if the initial state is not too high. So for $n_{osc} = 2$ one has $P_{n_i s \to (n_i \pm 1)p_1}^m / P_{n_i s \to 1s}^m < 1$ if n_i <1600 τ ⁴. The last conditions is well satisfied for available laser pulses $(\tau > 4$ a.u.) and engineered Rydberg states (n_i) < 600).

The SPA can also be used for a description of ultrashort laser pulse interactions in multielectron systems. Here we also assume that the applicability conditions (9) are satisfied. In the zeroth approximation this approach results in an expression for transition amplitudes similar to Eq. (17). As in the one-electron case the net displacement and momentum gained by each electron from the field are zero and one needs first-order corrections to describe the pulse action on the atom. Together with the effects considered previously these corrections include electron-electron correlation effects. So, for example, if the residual vector potential A_1 is zero, the electron-electron repulsion leads to terms of the form

$$
\int_{-\infty}^{\infty} dt t \frac{\partial}{\partial t} V_{ee}(|\mathbf{r}_i + \Delta \mathbf{R}(\boldsymbol{\eta}_i) - \mathbf{r}_j - \Delta \mathbf{R}(\boldsymbol{\eta}_j)|) \tag{47}
$$

in the first-order correction to $\hat{\Delta}$ ($\hat{S} = \hat{S}_0[\hat{I} + i\hat{\Delta}]$). In expression (47), r_i and r_j specify positions of the interacting electrons, $\eta_i = t - \alpha x_i$, and x_i is the *x* coordinate of vector r_i . The correlation terms are nonzero only if the laser field is spatially nonuniform. If the field characteristic size is comparable with the sizes of the initial orbital, one should take into account retardation effects. Nevertheless, the mechanism leading to the terms (47) is similar to the mechanism of the pulse-propagation effect $[18]$ and one can expect that these terms do not contribute significantly to the transition probabilities.

IV. SUMMARY AND CONCLUSIONS

The hydrogen atom interaction with an ultrashort electromagnetic pulse has been studied for pulse durations τ shorter than $1/\varepsilon_i$. The nonrelativistic sudden perturbation approximation has been used to calculate the initiated transition probabilities. The analytical expression for the evolution operator has been derived up to the first order of the approximation.

The case of real laser pulses is considered where the field does not change the initial electron position and momentum when the pulse action is complete. Such pulses do not affect hydrogen atoms in the zeroth order of the sudden perturbation approximation. The first order takes into account the effect of the magnetic field and also the change in the electron-nuclear interaction along the trajectory \mathbf{R}_i forced by the laser electric field. Both these effects are nonzero for real pulses. They cause magnetic and nonmagnetic transitions, respectively.

In this paper we considered only bound-bound transitions at the hydrogen-atom interaction with ultrashort laser pulses. Work on bound-free transitions is under consideration. The sudden perturbation approximation can also be applied to ultrashort-pulse interactions with multielectron systems (atoms and molecules). Presently, the results obtained can be used to study the Rydberg atom interaction with trains of intense ultrashort laser pulses which is of current interest. Also, we hope that the approach used will find more applications with the development and implementation of new methods for generation of shorter and more powerful pulses. Progress in this direction has already begun [43,44].

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