Coulomb-Volkov approach of atom ionization by intense and ultrashort laser pulses

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We present a nonperturbative theoretical approach, based on Coulomb-Volkov-type states, which is able to predict both angular and energy distributions of ejected electrons when atoms interact with a very short and intense laser pulse. In a previous paper [Eur. Phys. J. D **11**, 191 (2000)], it was shown that, for atomic hydrogen targets, this theory makes accurate predictions as long as the interaction time does not allow more than two optical cycles. Recently, multigigawatt laser pulses with a full width at half maximum of less than two optical cycles have been generated by Nisoli *et al.* [Opt. Lett. **22**, 522 (1997)] at $\lambda = 800$ nm. In the present paper, it is shown that predictions of the Coulomb-Volkov approach for the ionization of a hydrogen atom by laser pulses similar to the ones generated by Nisoli *et al.* are in very good agreement with the predictions of an ''exact'' numerical treatment. Further, the domain where the Coulomb-Volkov theory applies is marked out by means of a consistent accuracy parameter and by comparison with an ''exact'' numerical treatment. It is shown that, subject to the above-mentioned condition, good predictions may always be issued as long as the interaction time does not exceed half the initial orbital period of the electron. For a given laser pulse duration, predictions are all the better that the laser field amplitude is high and the initial quantum number is large.

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I. INTRODUCTION

In a previous paper, hereafter referred to as paper I, we introduced a simple nonperturbative approach of atom ionization by intense and ultrashort laser pulses based on Coulomb-Volkov (CV) states [1]. A preliminary study showed that it provides accurate predictions as long as the laser field does not perform more than two oscillations. In addition, there is every indication that the pulse duration should not exceed half the period of the initial orbital of the ejected electron. It is worth noting that the two conditions do not restrict the application of the CV theory since multigigawatt, 4.5-fs (full width at half maximum), 800 nm laser pulses have been achieved in Vienna [2]. Thus each pulse exhibits less than two oscillations, and ionization predictions can be made by the present CV theory for target orbitals with a principal quantum number $n \geqslant 4$. Of course, similar predictions could be made with a full numerical treatment of the time-dependent Schrödinger equation (TDSE). Nevertheless, TDSE calculations often require both long CPU time and large memory size. Further, femtosecond terawatt lasers have been developed and intensities as high as 10^{18} W cm^{-2}, or even higher, have been reached. Under these conditions, TDSE calculations exhibit numerical difficulties (see results hereafter). Furthermore, previous simple models already developed for tunnel or barrier suppression ionization (see, e.g., Ref. [11]) appear not to be well adapted to very short laser pulses (see the discussion in Sec. III D).

When atoms of gas or small clusters are illuminated by such short and intense laser pulses, a highly ionized plasma is created in a time much shorter than the pulse length [3,4]. Actually, ionization is completed after the first half cycle of a 3.5×10^{18} W cm^{-2}, 20 fs, 800 nm laser pulse [1] (see results hereafter). Under such conditions, total ionization takes place in a few atomic units of time, and the plasma may be created in a comparable period of time. Its further evolution may be described by a particle-in-cell code, which usually starts with a grid uniformly filled by a Maxwellian distribution of an ensemble of electrons [4]. In this procedure, the plasma is assumed to be thermalized at a given temperature from the very beginning, but it is not clear whether the initial electron energy distribution is Maxwellian.

Therefore, there is a need for a simple and reliable method to predict electron distributions for high intensities and very short interaction times. The CV method introduced in paper I is a good candidate. It is simply based on a good approximation of the exact wave function of a hydrogenlike atom merged in an external time-dependent electric field. This approximate wave function, which is called a CV state [5], is all the more accurate that the influence of the nuclear field during the pulse is negligible. The aim of the present paper is to give a more detailed derivation of this, and to introduce consistent criteria which indicate where the CV method applies. Angular distributions are also investigated here, as well as energy distributions already considered in paper I. Again, CV predictions are compared with TDSE calculations when the latters can be performed. Atomic units are used throughout unless otherwise stated.

II. THEORY

A. Description of the laser pulse

Our study is made for a pulsed laser, whose field is linearly polarized along the z axis, and whose envelope contains less than two oscillations. The finite pulse duration is featured through a sine-square envelope. Thus the field expression is

$$\vec{F}(t) = \begin{cases} \vec{F}_0 \sin(\omega t + \varphi) \sin^2\left(\dfrac{\pi t}{\tau}\right) & \text{if } 0 < t < \tau \\ \vec{0} \quad \text{elsewhere,} \end{cases} \tag{2.1}$$

where τ is the duration of the pulse. As in paper I, all calculations are made with a time-symmetric pulse, which implies

$\varphi=-(\omega\tau/2)+(\pi/2)$. The photon energy is set to $\hbar\omega=0.05$ a.u. to overlap the energy range of photons commonly generated by Ti:sapphire lasers. The electric field of the laser is derived from a vector potential $\vec{A}(t)$, i.e.:

$$\vec{F}(t)=-\frac{\partial\vec{A}(t)}{\partial t}. \tag{2.2}$$

The vector potential is

$$\vec{A}(t)=\vec{A}(t_0)-\int_{t_0}^{t}dt'\vec{F}(t'), \tag{2.3}$$

where t_0 is the time when the laser-atom interaction begins, and $\vec{A}(t_0)$ is an arbitrary constant value of the vector potential before any interaction. To fulfill asymptotic conditions on the wave function of the system, in what follows, one must set $\vec{A}(t_0)=\vec{0}$. Thus, if the field makes many oscillations within the sine-square envelope, it is easy to show that $\vec{A}(\tau)$ is all the closer to zero that the number of oscillations is large (it is true only because the pulse duration is finite): this is the domain where ionization is dominated by multiphoton absorption. Conversely, if the pulse is so short that the field has no time to oscillate, the magnitude of $\vec{A}(\tau)$ may be high. In this case, it was shown that the laser field does not exhibit any quantum aspect; it acts as a pure classical field [10] and the ionization either by tunnel effect or, for strong enough fields, by barrier suppression ionization (BSI) are the dominant mechanisms.

B. Time-dependent calculations

The results obtained with the present theory are compared to the ''exact'' solution in order to validate the method and to define the domain of parameters where it applies. In this section, we briefly describe the technique implemented to solve numerically the time-dependent Schrödinger equation:

$$i\frac{\partial\Psi(\vec{r},t)}{\partial t}=\left(-\frac{\vec{\nabla}^2}{2}-\frac{Z}{r}+\vec{r}\cdot\vec{F}(t)\right)\Psi(\vec{r},t). \tag{2.4}$$

This method was developed several years ago. It was designed more specifically to compute above threshold ionization spectra of electrons emitted during the interaction between an atom and a strong laser pulse [6]. However, its application to the present case is straightforward. The Schrödinger equation is solved by expanding the solution on basis functions built with products of *B*-spline functions for the radial coordinate, and of spherical harmonics for the angular coordinates. Thus, for a linearly polarized laser field, the solution reads

$$\Psi(\vec{r},t)=\sum_i a_i(t)\frac{B_i^k(r)}{r}Y_l^0(\theta,\varphi), \tag{2.5}$$

where $B_i^k(r)$ is the ith *B*-spline function of order k. In Eq. (2.5), m is set to zero because the initial orbital is the $1s$ state and the laser is linearly polarized. *B*-spline functions are square-integrable local functions that are defined by dividing the radial axis (r goes from 0 to $R_{\max}$) in contiguous intervals with particular continuity conditions at their boundaries [6]. The efficiency of *B* splines has been proven in atomic and molecular physics as well as in studies of the dynamics of laser-atom interactions. It is a consequence of remarkably useful numerical properties [6]. Substituting expression (2.5) for Ψ into Eq. (2.4) leads to a set of differential equations for the coefficients $a_i(t)$. These equations are solved by an implicit scheme accounting for the fact that due to the locality of *B*-spline functions, the matrices have a band structure whose width is $2k-1$. Once the coefficients are known at the end of the pulse, the emitted electron spectrum is obtained by projection on continuum eigenstates of the field-free atomic system. Similarly, angular distributions can be obtained by projection onto the eigenstate, which represents an outgoing electron in the nuclear field with a momentum pointing in a particular direction.

The intrinsic symmetry of the atom naturally leads us to express the wave functions in spherical coordinates. However, in the present case of extremely short and intense laser pulses, the spherical symmetry is broken because the system is now dominated by a strong field. Therefore, its symmetry is closer to the cylindrical symmetry imposed by a linearly polarized field. As a consequence, the expansion in terms of angular momenta may blow up, thus setting an upper limit for the applicability of this ''exact'' approach, and therefore justifying the search for specifically adapted methods.

C. CV theory

The Coulomb-Volkov approach was introduced in paper I [1]. In this paper, we concluded that the CV theory is reliable as long as the influence of the nuclear field on the variation of the electron energy during the laser pulse is smaller than the effect of the laser field itself. We will now show that this physical hypothesis is necessary to derive an analytical form of the CV state which describes the evolution of an electron both in the Coulomb potential and in the laser field. Therefore, we expect our theory to give reliable predictions as long as this assumption is true: that is, with intense enough laser fields. We start from the time-dependent Schrödinger equation. We use the dipole approximation in the length gauge. In this context, the laser-atom interaction W is

$$W=-\vec{D}\cdot\vec{F}(t), \tag{2.6}$$

where $\vec{D}=q\vec{r}=-\vec{r}$ is the dipole associated with the one-electron atom. Hence the Schrödinger equation may be written as

$$i\frac{\partial\Psi(\vec{r},t)}{\partial t}=\left(-\frac{\vec{\nabla}^2}{2}-\frac{Z}{r}+\vec{r}\cdot\vec{F}(t)\right)\Psi(\vec{r},t), \tag{2.7}$$

where Z is the nuclear charge of the atom. In order to find an approximate solution of Eq. (2.7) with an ultrashort and ultraintense laser pulse, let us first consider the state of a free electron in an oscillating electric field. This is a well-known

Volkov state $\mathcal{V}(\vec{r},t)$, which is an exact solution of Eq. (2.7) with $Z=0$. Explicitly, it reads

$$\mathcal{V}(\vec{r},t)=\exp\left(i\vec{p}(t)\cdot\vec{r}-\frac{i}{2}\int_0^t dt' p^2(t')\right), \tag{2.8}$$

with

$$\vec{p}(t)=\vec{k}+\vec{A}(t), \tag{2.9}$$

where $\vec{k}$ is the electron momentum in the absence of an electric field. For ultraintense laser pulses, i.e., lasers with an electric-field amplitude comparable to or even higher than the Coulomb field experienced by an electron in the ground state of hydrogen, the Volkov state must be close to the exact solution of Schrödinger equation (2.7). Then it is reasonable to feature the most rapid oscillations of $\Psi(\vec{r},t)$ with time by means of a factor similar to the Volkov phase. Thus let us write the *ingoing* scattering wave function as

$$\Psi^-(\vec{r},t)=f^-(\vec{r},t)\exp\left(i\vec{p}(t)\cdot\vec{r}-\frac{i}{2}\int_\tau^t dt' p^2(t')\right), \tag{2.10}$$

where $\vec{p}(t)$ is a time-dependent momentum to be defined. Substituting expression (2.10) for $\Psi(\vec{r},t)$ into Eq. (2.7) leads to the equation for $f^-(\vec{r},t)$:

$$i\frac{\partial f^-(\vec{r},t)}{\partial t}=-\frac{\vec{\nabla}^2}{2}f^-(\vec{r},t)-i\vec{p}(t)\cdot\vec{\nabla}f^-(\vec{r},t)+\left[-\frac{Z}{r}+\left(\frac{\partial\vec{p}(t)}{\partial t}+\vec{F}(t)\right)\cdot\vec{r}\right]f^-(\vec{r},t). \tag{2.11}$$

Following a step similar to Presnyakov and co-workers [7,8], we assume that the change in the electron motion is mainly due to the external electric field. Therefore, one sets

$$\frac{\partial\vec{p}(t)}{\partial t}=-\vec{F}(t), \tag{2.12}$$

which implies

$$\begin{aligned}&\vec{p}(t)=\vec{k}+\vec{A}^-(t)=\vec{k}-\int_\tau^t dt'\vec{F}(t') \quad \text{if } t<\tau\\ &\vec{p}(t)=\vec{k} \quad \text{if } t\geqslant\tau,\end{aligned} \tag{2.13}$$

where $\vec{A}^-(t)=-\int_\tau^t dt'\vec{F}(t')$ is specifically related to the ingoing wave function. Thus we find a time-dependent momentum similar to Volkov's. It is worth noting that Eq. (2.12) is nothing but the fundamental principle of classical dynamics in which the force due to the Coulomb field is neglected during the interaction. Now, with Eq. (2.12), Eq. (2.11) reduces to

$$i\frac{\partial f^-(\vec{r},t)}{\partial t}=\left[-\frac{\vec{\nabla}^2}{2}-i\vec{p}(t)\cdot\vec{\nabla}-\frac{Z}{r}\right]f^-(\vec{r},t). \tag{2.14}$$

In agreement with Eq. (2.12), it is reasonable to assume that the small time variation of $f^-(\vec{r},t)$ is mainly due to the external electric field. Since the second term on the right-hand side of Eq. (2.14) is the only one which depends on the field, one may split this equation as follows:

$$i\frac{\partial f^-(\vec{r},t)}{\partial t}=-i\vec{A}^-(t)\cdot\vec{\nabla}f^-(\vec{r},t), \tag{2.15}$$

$$\left(-\frac{\vec{\nabla}^2}{2}-i\vec{k}\cdot\vec{\nabla}-\frac{Z}{r}\right)f^-(\vec{r},t)=0. \tag{2.16}$$

Indeed, the solution of this new system implies that $f^-(\vec{r},t)$ is a particular solution of Eq. (2.14). First, for $t>\tau$, one has $\vec{A}^-(t)=\vec{0}$, and, from Eq. (2.15) $f^-(\vec{r},t)$ is time independent; let us call it $f_0(\vec{r})$. It is clearly an unperturbed function because the laser no longer interacts with the atom. Since operators in Eq. (2.16) do not depend on time, it should be an equation for a stationary state. Hence it appears reasonable that $f_0(\vec{r})$ is determined by Eq. (2.16). The solution of the latter equation is a confluent hypergeometric function like

$$f_0(\vec{r})=\frac{1}{(2\pi)^{3/2}}\exp(\pi\nu/2)\Gamma(1+i\nu)F_1(-i\nu,1,-ikr-i\vec{k}\cdot\vec{r}), \tag{2.17}$$

with $\nu=Z/k$. Now, if $t<\tau$, Eq. (2.15) may be integrated formally as

$$f^-(\vec{r},t)=f_0(\vec{r})-\int_\tau^t dt'\vec{A}^-(t')\cdot\vec{\nabla}f^-(\vec{r},t'). \tag{2.18}$$

If the interaction time is short enough, we may consider that the integral term in Eq. (2.18) is close to zero and $f_0(\vec{r})$ should be a good approximation of $f^-(\vec{r},t)$. Under these conditions, we can rewrite the approximate solution of the time-dependent Schrödinger equation (2.7) as

$$\begin{aligned}\Psi^-(\vec{r},t)\simeq\chi_f^-(\vec{r},t)&=f_0(\vec{r})\exp\left(i\vec{p}(t)\cdot\vec{r}-\frac{i}{2}\int_\tau^t dt' p^2(t')\right)\\ &=f_0(\vec{r})\mathcal{L}^-(\vec{r},t),\end{aligned} \tag{2.19}$$

where $\chi_f^-(\vec{r},t)$ is called the incoming CV wave function [5]. It is worth noting that the CV state [Eq. (2.19)] becomes a standard Volkov state when the nuclear charge is zero because $f_0(\vec{r})=1$. By analogy, we can define a CV state for an outgoing wave function, starting from a bound wave function (see Appendix A). It reads

$$\begin{aligned}\chi_i^+(\vec{r},t)&=\varphi_i(\vec{r},t)\exp\left(i\vec{A}^+(t)\cdot\vec{r}-\frac{i}{2}\int_0^t dt' A^{+2}(t')\right)\\ &=\varphi_i(\vec{r},t)\mathcal{L}^+(\vec{r},t),\end{aligned} \tag{2.20}$$

where $\varphi_i(\vec{r},t)$ is the initial unperturbed bound state. The vector potential $\vec{A}^+(t)=-\int_0^t dt'\vec{F}(t')$ in Eq. (2.20) is specifically related to an outgoing wave function. It is easy to show that the level of approximation for this outgoing state is equivalent to the one for the ingoing CV state. Indeed, the substitution of either the incoming or the outgoing CV states for Ψ in the Schrödinger equation leads to similar residual terms, i.e.,

$$\left(H-i\frac{\partial}{\partial t}\right)\chi^-=\vec{\nabla}f_0\cdot\vec{\nabla}\mathcal{L}^-, \tag{2.21}$$

$$\left(H-i\frac{\partial}{\partial t}\right)\chi^+=\vec{\nabla}\varphi_i\cdot\vec{\nabla}\mathcal{L}^+. \tag{2.22}$$

This implies that the accuracy of a CV calculation is equivalent in both the outgoing and the ingoing approaches. An evaluation of this accuracy is made in Sec. II E.

D. Ionization probability

Although it is important to know the total ionization probability, energy, and angular distributions of the ejected electrons provide both more physical insight and useful data for applications. They require to determine the transition amplitude T_{fi}. A simple way to calculate T_{fi} is to project the outgoing total wave function after the interaction with the electric laser field onto the final continuum state. The final state is a continuum Coulomb wave function $\mathcal{C}_{\vec{k}}^-$. For short enough pulses, a CV state $\chi_i^+(\vec{r},t)$ should be a good approximation of the total electronic wave function during the laser-atom interaction. Under these considerations, the transition amplitude is

$$T_{fi}=\lim_{t\to+\infty}\langle\mathcal{C}_{\vec{k}}^-|\chi_i^+\rangle. \tag{2.23}$$

It is easily shown that this scalar product is not changed by the unitary transformation into the velocity gauge. It only means that the gauge invariance cannot be invoked as a test in the present case. Then, the energy distribution of the ejected electrons $\partial P^{CV}/\partial E_k$ is simply

$$\frac{\partial P^{CV}}{\partial E_k}=k\int d\Omega_k|T_{fi}|^2, \tag{2.24}$$

where k appears in place of k^2 because the wave functions are normalized in the momentum representation. The angular distribution for a given energy is

$$\frac{\partial^2 P^{CV}}{\partial E_k\partial\Omega_k}=k|T_{fi}|^2, \tag{2.25}$$

where the integral over the azimuth angle φ is 2π because of the symmetry of the problem with a laser pulse linearly polarized along the z axis.

E. Accuracy of the CV theory

The approximate solution of the Schrödinger equation obtained in Sec. II C assumes a weak influence of the nuclear field during the pulse, which leads to Eq. (2.12). Therefore, it is reasonable to think that the higher the laser field amplitude, the better the approximation. A first way to quantify the accuracy of CV predictions is to compare it directly to TDSE predictions, that are considered as exact ones in the domain where convergence of TDSE calculation is workable. It is achieved by the definition of a relative accuracy parameter ϵ which is based on CV and TDSE electron energy spectra, i.e.,

$$\epsilon=\frac{\displaystyle\int_0^{+\infty}dE_k\left|\frac{\partial P^{TDSE}}{\partial E_k}-\frac{\partial P^{CV}}{\partial E_k}\right|}{\displaystyle\int_0^{+\infty}dE_k\frac{\partial P^{TDSE}}{\partial E_k}}. \tag{2.26}$$

Since $\partial P^\alpha/\partial E_k\geqslant 0$ (where α is either TDSE or CV), it is clear that ϵ is a severe test of the reliability of the CV method. Then, it appears interesting to plot this error as a function of F_0 for different nuclear charges Z in order to first exhibit the influence of the nuclear field. The error is evaluated numerically from the CV and TDSE spectra in Sec. III.

We also introduce a self-consistent appreciation of the present CV theory. This consists of seeking for the error made in replacing $f^-(\vec{r},t)$ by $f_0(\vec{r})$ in Eq. (2.19). In view of Eq. (2.18), it is reasonable to compare $f_0(\vec{r})$ with $\int_\tau^t dt'\vec{A}^-(t')\cdot\vec{\nabla}f^-(\vec{r},t')$. To do this, we define the analytical relative error $\mathfrak{E}$ made in the Coulomb-Volkov wave function, when we do not take into consideration the second term in Eq. (2.18), as (see Appendix B):

$$\mathfrak{E}=k\beta(k)\frac{\tau}{6}, \tag{2.27}$$

with

$$\beta(k)=\left|\frac{\langle\partial f_0/\partial z\rangle}{\langle f_0\rangle}\right|. \tag{2.28}$$

$\mathfrak{E}\ll 1$ implies $f^-(\vec{r},t)\simeq f_0(\vec{r})$.

III. RESULTS AND DISCUSSION

A. Ionization during the pulse rise

A TDSE investigation of the interaction of a H($1s$) target with a realistic laser pulse characterized by 3×10^{18} W cm^{-2}, 20 fs, 800 nm, show that ionization is completed at the beginning of the pulse. Although calculations are not fully converged, results are good enough to ensure that ionization is completed after the first half cycle. In what follows, we exhibit the limitations of a full numerical treatment when the intensity of the laser pulse becomes too high. In Fig. 1, the time evolution of the total electronic population initially in the ground state of hydrogen is displayed for each angular momentum. It is clear that the atom is almost fully ionized at $t=60$ a.u. The period corresponding to the wavelength is about 114 a.u. Thus, within half a cycle, the atom is entirely brought into the continuum.

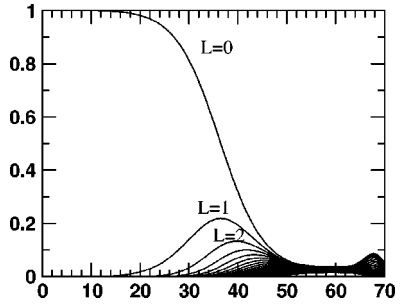

FIG. 1. Ionization of H($1s$) by a 20-fs laser pulse whose peak intensity is 3×10^{18} W cm^{-2} at a wavelength of 800 nm. The figure shows the time evolution of the population of each angular momentum as predicted by the TDSE code within the first half-cycle of the pulse.

Let us now consider Fig. 2, which displays the same curves as in Fig. 1, but with a logarithmic scale on the vertical axis. For $t>70$ a.u., all populations tend to stabilize around approximately the same value, although there is no physical reason for it. Further, with a laser pulse characterized by 3×10^{13} W cm^{-2}, 24.8 fs, and $\omega=2.0$ eV, Cormier and Lambropoulos [6] predicted that, at any time, the L population decreases when L increases in the velocity gauge, whereas calculations in the length gauge give results similar to Fig. 2, a behavior typical to a failure to converge. This fact is noteworthy when the ionization probability is close to unity. Further, more than 100 angular momenta are often necessary to obtain a stabilization of the energy distribution of the ejected electrons. This is due to the breakdown in the spherical symmetry when the electric field is too high. In fact, we will show that, in this case, the loss of spherical symmetry causes the electrons to eject principally along the polarization direction. Therefore, cylindrical coordinates would probably be more adapted to perform full numerical calculations. Note that calculations in the velocity gauge are extremely cumbersome in the case of a very intense field, because the propagation algorithm, which is based on an iterative scheme, then requires too many iterations. In this case, a more adapted theory is needed to predict angular distributions and energy spectra. A good candidate is the present CV approach, because it is especially designed for intense external fields. Since this simple analytical approach is established with some approximations, it is necessary to

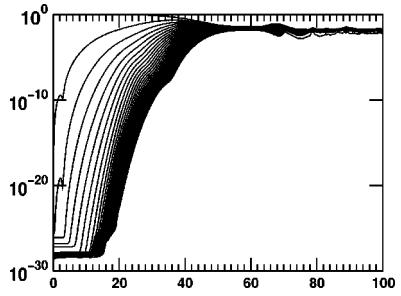

FIG. 2. Same as in Fig. 1 with a logarithmic scale on the vertical axis. All populations tend to stabilize around approximately the same value, because the code does not converge (see text).

mark out the domain where it applies by looking at the expected accuracy of predictions. This is the aim of Sec. III B.

B. Evaluation of the accuracy of the CV method

First, let us recall that the CV theory pertains to the sudden approximation. This implies that results must be valid as long as the laser pulse duration is a given part of the initial orbital period $T_{n,Z}$, which is equal to $T_{n,Z}=2\pi n^3/Z^2$ in Bohr's model. The evolution of the CV accuracy ϵ (as defined in Sec. II E), with a laser field amplitude F_0 reported in Fig. 3, shows that it is the case. The first column is for $Z=1$, the second for $Z=2$ and the third for $Z=3$. Rows correspond to different laser pulse durations τ. In all cases, τ is proportional to an orbital period which depends only on Z for a given state of hydrogen (it is the ground state here). $\tau=T_{1,Z}/4$, $T_{1,Z}/2$, and $T_{1,Z}$ for the first, second, and third rows, respectively. The shortest pulse duration is $\tau=T_{1,3}/4\approx0.17$ a.u. ($Z=3$). Of course, this is a very short time, but one must keep in mind that our aim is to look for the accuracy of the CV approach with respect to the orbital period. Moreover, by scaling $T_{n,Z}$, it is possible to extend the present conclusions to longer periods of the initial orbital, i.e., to laser pulses of a few femtoseconds. Further, it is worth noting that the sudden approximation appears applicable for the smallest values of the field amplitude because ϵ takes the same value in a row, i.e., for a pulse duration which is a given fraction of the orbital period. In paper I, we concluded that good enough predictions are obtained as long as $\tau\leqslant T_{n,Z}/2$. We are now in a position to quantify this statement: as shown by the second row of Fig. 3, $\tau\leqslant T_{n,Z}/2$ corresponds to a 20% accuracy. In all applications, this limit will be taken as a criterion which delimits the region where CV applies.

Now, plotting ϵ as a function of the laser field amplitude for different nuclear charges Z allows us to exhibit the influence of the Coulomb potential on the accuracy of the CV method. It is worth recalling that its effect is ignored in the dynamics of the process as stated in Eq. (2.12). In Fig. 3, it appears that ϵ begins to decrease for a given value of F_0 called F_{0c}. We stop plotting ϵ when the TDSE code no longer converges. This is the case for very large values of F_0 for which the ionization is completed. Indeed, one would expect that ϵ would go to zero when the laser field continues to increase, because the effect of the nucleus field on the dynamics of the ionization becomes less and less important. However, due to uncertainties in TDSE calculations when F is too high, ϵ stops decreasing and begins to increase beyond a large value of F where TDSE calculations can still be performed, but without any guarantee of the accuracy. In fact, for such values of F, the predictions of the CV theory become much more reliable than any other prediction. Therefore, $F\geqslant F_{0c}$ is the favorite domain of the CV theory. F_{0c} is the value of F_0 beyond which Eq. (2.12) becomes meaningful, because the effect of the nuclear field appears very weak compared to the laser field. Therefore, in this domain of laser intensities, the CV theory applies for interaction times longer than the ones required by the previous sudden approximation criterion. It is no wonder, since at high laser intensities

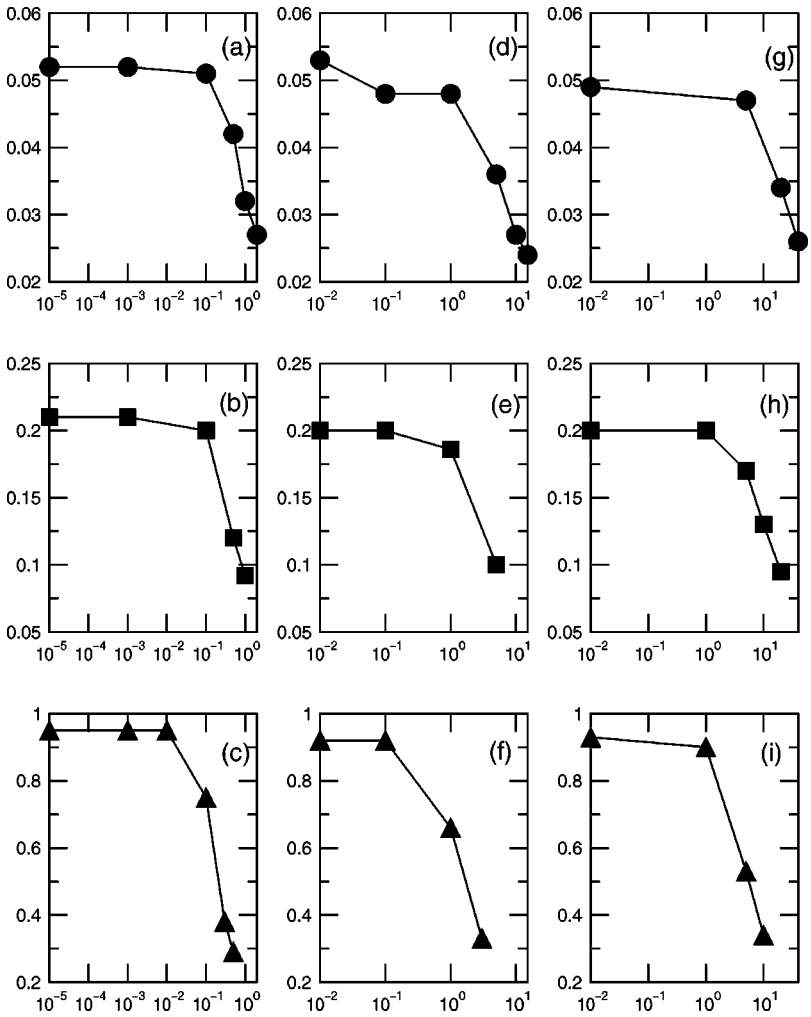

FIG. 3. For the ionization of H($1s$), the accuracy (see text) is plotted as a function of the laser field amplitude F_o for various nuclear charges and various pulse durations. First column: $Z=1$. Second column: $Z=2$. Third column: $Z=3$. First row: $\tau=T_{1,Z}/4$. Second row: $\tau=T_{1,Z}/2$. Third row: $\tau=T_{1,Z}$.

ionization is completed slightly after the beginning of the laser pulse, and the ejected electrons are rapidly driven away from the nucleus by the laser field. Then statement (2.12) becomes more realistic for the rest of the pulse, which means that the further evolution of the electron state is described accurately by the CV state.

In all cases, the total ionization probability is close to 10^{-2} when $F_0=F_{0c}$. Let us examine F_{0c}. To do this, in Fig. 4 we plot both the total ionization probability and ϵ as functions of F_0 for $\tau=T_{1,Z}/2$ (the domain where the CV method applies whatever the laser field amplitude), again for $Z=1$, 2, and 3 [Figs. 4(a), 4(b), and 4(c), respectively]. One notes that the probability increases very rapidly beyond F_{0c}. Therefore, F_{0c} demarcates perturbative ($F_0 \leqslant F_{0c}$) and nonperturbative ($F_0 > F_{0c}$) regimes. It is worth noting that $F_{0c} \approx 2F_{crit}$, where $F_{crit}=Z^3/16$ is the critical field in the saddle point model; F_{crit} lowers the Coulomb barrier to the energy of the bound state $1s$, thus allowing the electron to escape classically. Hence F_{0c} points out the value of the field for which the transparency of the barrier becomes effective.

Now let us look for the critical energy E_{tc}, which has to be transferred to the electron to consider that the laser field influence is noticeable compared to the nuclear field, i.e., the energy which corresponds to the upper limit of the perturbation region. From Eq. (B6) in Appendix B, it is easy to show that $E_{tc} \simeq F_{0c}^2 \tau^2/8$. Taking F_{0c} as determined above for an interaction time $T_{1,Z}/2$, the critical energy transfer E_{tc} is

$$E_{tc} \simeq \frac{|E_I|}{26}, \tag{3.1}$$

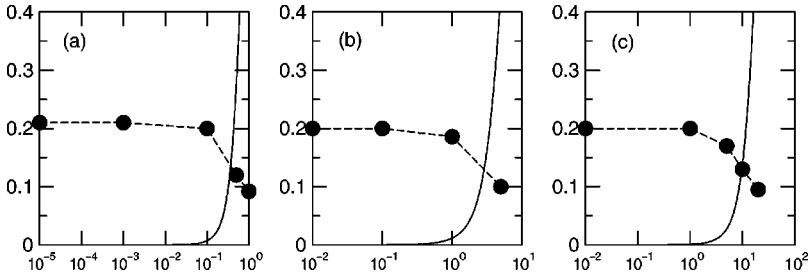

FIG. 4. For the ionization of H($1s$), ϵ (dashed line) and the total ionization probability P (full line) are plotted as functions of the laser field amplitude F_o. The pulse length is $\tau=T_{1,1}/2$. (a) $Z=1$, (b) $Z=2$, and (c) $Z=3$. Figures show that ϵ improves above the value of F_o where P becomes appreciable.

TABLE I. Value of the parameter β (see text) as a function of the ejected electron energy E_k and of the initial state.

E_k	10 a.u.	30 a.u.	100 a.u.
H(1s)	0.21	0.1	0.4×10^{-1}
H(2s)	0.27×10^{-1}	0.13×10^{-1}	0.52×10^{-2}
H(3s)	0.73×10^{-2}	0.34×10^{-2}	0.14×10^{-2}

where E_I is the ionization potential. Thus an energy transfer of only 4% or so of the binding energy is enough to leave the perturbation regime.

Finally, it is worth pointing out that, in the perturbation regime ($F_0 \leqslant F_{0c}$), ϵ takes a constant value. Indeed, both CV and TDSE energy distributions are proportional to the laser intensity in this regime. Since $\epsilon < 20\%$ as long as $\tau \leqslant T_{n,Z}/2$, one may conclude that CV theory also gives reliable predictions in the perturbation range.

In the above discussions the numerical accuracy of the CV theory was investigated by comparing its predictions directly with TDSE ones. Now it appears interesting to study the analytical error coming from the derivation of the CV wave function itself. We already pointed out that this depends on the parameter β [Eqs. (2.27) and (2.28) in Sec. II E], which is both a function of the final momentum of the electron and a functional of the initial state. Although no analytical expression of β can be found, it may be shown that it tends to a constant when k goes to zero. In fact, it is more interesting to know its behavior for a large energy transfer where CV better applies. We summarize some values of this parameter in Table I. High laser intensities produce high ejection energies. In this case, β may be approximated by the analytical expression

$$\beta(k,n) \simeq \frac{2}{k^{3/2}n^3}, \tag{3.2}$$

where $k \simeq F_0\tau/2$ [see Appendix B, Eq. (B6)]. Substituting Eq. (3.2) for β into Eq. (2.27) leads to the analytical error roughly proportional to

$$\mathfrak{E} \propto \frac{\sqrt{\tau}}{n^3\sqrt{F_0}}. \tag{3.3}$$

This value of the accuracy stems from the hypothesis $f = f_0$ in Sec. II. It cannot be compared with ϵ because, first, it represents only the error at the maximum of the energy distribution and, second, it predicts the accuracy for large values of the ejected electron energy where the TDSE code does not converge, i.e., in a region where ϵ cannot be calculated. Therefore, expression (3.3) of $\mathfrak{E}$ provides us with useful information. As expected, the higher the laser field amplitude F_0, the smaller $\mathfrak{E}$. It gives additional indication that the CV theory is well adapted to the non-perturbative regime. Further, it shows that predictions are all the better that the initial quantum number is large.

C. Energy and angular spectra

In paper I, we showed that for high enough laser intensities the ionization is completed in the first half-cycle of the pulse. Therefore, it is useful to study fields which do not oscillate. In this case, the field exhibits essentially a classical aspect and the energy transferred to the atom reads

$$E_t = \frac{1}{2}\left| -\int_0^\tau dt F(t)\right|^2 = \frac{\vec{A}(\tau)^2}{2}. \tag{3.4}$$

When E_t is greater than $|E_I|$, where $E_I < 0$ is the initial binding energy of the electron, it can be shown analytically that the CV spectrum peaks at an energy E_{peak} [9] given by

$$E_{peak} = E_t + E_I. \tag{3.5}$$

We also showed in paper I that $T_{fi} \to 0$ when the field makes a few oscillations within the pulse envelope. As a consequence, our method applies only when the field makes less than 1.7 oscillation. In addition, we stated that accurate predictions with the CV approach could be obtained when $\tau \leqslant T_{n,Z}/2$, a condition related to the sudden approximation. Under these conditions, the CV theory works very well. In Sec. III B, we indicated that the latter inequality corresponds to an accuracy ϵ better than 20% according to our definition of ϵ, which is a fairly hard criterion. In fact, a qualitative agreement is still obtained for a pulse duration equivalent to the orbital period and it is all the better that the laser intensity is high.

Now let us address the angular and energy distributions predicted by CV theory when the interaction time is small enough to guarantee that the sudden approximation applies. For the sake of simplicity, we are dealing with targets of atomic hydrogen in their ground state. The interaction time which corresponds to a half-orbital period is $\tau = 3.14$ a.u. Although this is a very short time, it is worth emphasizing that the forthcoming analysis remains valid for much longer interactions times when higher orbitals are considered. For instance, the orbital period of the valence electron 5s of rubidium is close to 100 a.u. ($\approx$2.5 fs).

Energy distributions are displayed in Fig. 5 for various amplitudes of the laser field. As expected, a very good agreement is found both in perturbative and nonperturbative regimes. Let us now look at angular distributions. They are displayed in Fig. 6 for the same value of the laser field amplitude as in Fig. 5. At least two values of the ejected electron energy are considered: one is close to the maximum of the distribution, whereas the other one is away from it. Various features may be pointed out.

(i) For an ejection energy close to the maximum of the energy distribution, a very good agreement is found between CV and TDSE angular distributions for all laser field amplitudes [also see Figs. 5(a)–5(d)].

(ii) For an ejection energy well above the maximum, CV and TDSE angular distributions show similar behaviors with comparable orders of magnitude. The quantitative agreement is all the better that the amplitude of the field is high, except in Fig. 6(g) ($F_0 = 2.0$ a.u. and $E_k = 1.0$ a.u.). In this case, the maximum of the energy distribution shows up at 4.2 a.u., i.e.,

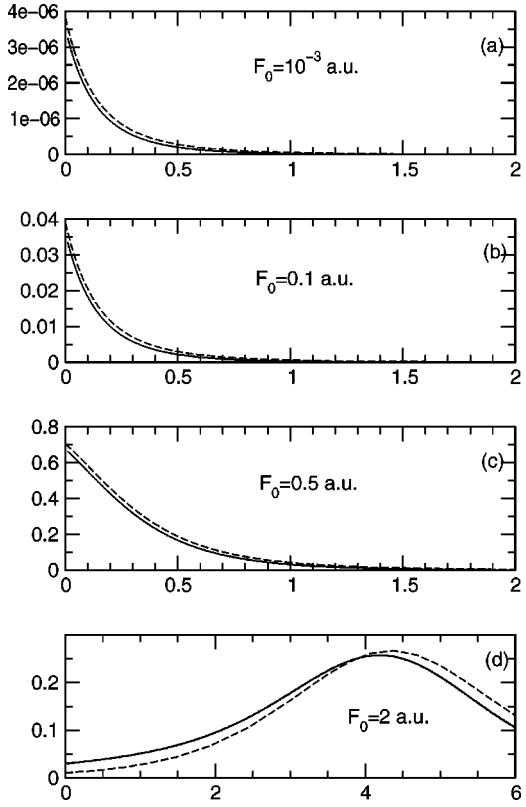

FIG. 5. Electron energy spectra predicted by TDSE (full line) and CV calculations (dashed line) with H(1*s*) targets for $F_o = 10^{-3}$ a.u. (a), 0.1 a.u. (b), 0.5 a.u. (c), and 2 a.u. (d). The interaction time is $\tau = 3.14$ a.u., and the photon energy is $\omega = 0.05$ a.u.

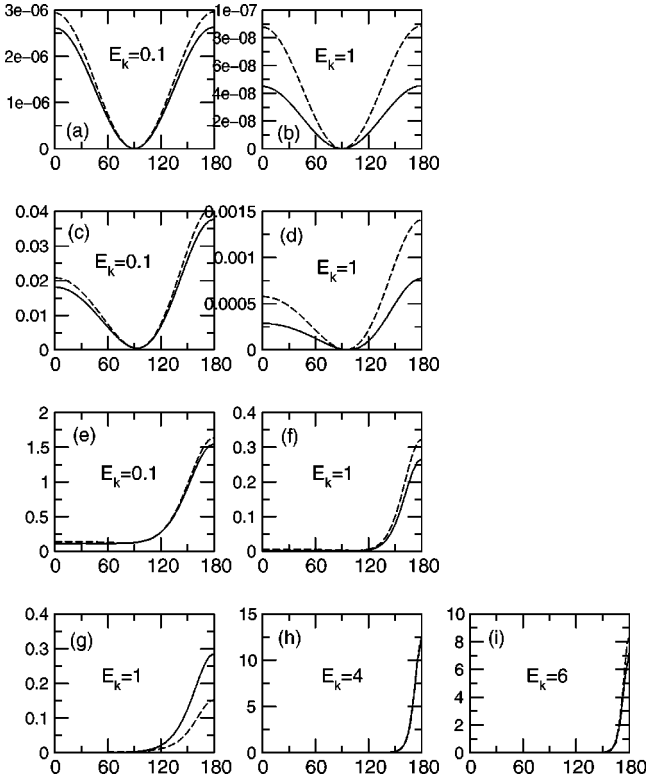

FIG. 6. Angular distributions of the ejected electron by TDSE (full line) and CV calculations (dashed line) with H(1*s*) targets. First row: $F_o = 10^{-3}$ a.u. Second row: $F_o = 0.1$ a.u. Third row: $F_o = 0.5$ a.u. Fourth row: $F_o = 2$ a.u. The interaction time is $\tau = 3.14$ a.u., and the photon energy is $\omega = 0.05$ a.u.

above the energy under consideration [see Fig. 5(d)]. Further, Fig. 5(d) shows that the TDSE probability at the ionization threshold is twice as much as the CV one. Although the total ionization probability is 1, it indicates that intermediate Rydberg states might have some influence on the ionization process.

(iii) For weak-field intensities, the angular distribution is symmetrical about 90°; there are as many electrons ejected in the direction of the polarization as in the reverse direction. Conversely, for high values of the laser field, electrons are ejected in a direction preferentially opposite to the polarization. This indicates that the influence of the initial electron distribution on the target becomes negligible, as if the electron were initially a free electron at rest in the frame of the target.

Figures 7(a) and 7(b) exhibit the influence of the Coulomb field of the nucleus when it is comparable to the laser field. Laser parameters are $F_0 = 1$ a.u., $\tau = 5$ a.u. [Fig. 7(a)], $\tau = 10$ a.u. [Fig. 7(b)], and $\omega = 0.05$ a.u. A perfect agreement is obtained for short enough pulses, i.e., for $\tau \leqslant 3$ a.u. We already indicated that the CV theory discards the influence of the Coulomb field of the nucleus on the electron during the laser-atom interaction. Long enough laser pulses allows the nucleus to slow down the electron; as a result, the energy distribution predicted by CV theory is shifted toward higher energies compared to TDSE predictions. This is illustrated by comparing Figs. 7(a) and 7(b).

Finally, Fig. 8 displays the density of probability for electrons ejected by a 4 fs, 800 nm, $3.5 \times 10^{14}\,\mathrm{W\,cm^{-2}}$ laser pulse from a hydrogen target in the 4*s* state. Again, the total ionization probability is close to unity. This features the ionization that could be achieved with laser pulses comparable to the ones generated by Nisoli *et al.* [2]. Thus it is clear that our present approach also applies to realistic laser pulses. For this very short pulse, the field does not oscillate much (1.6 oscillations with the present laser features), and the photon energy is not well defined. In fact, Nisoli *et al.* showed that the spectral band is very large, and appears as a white spec-

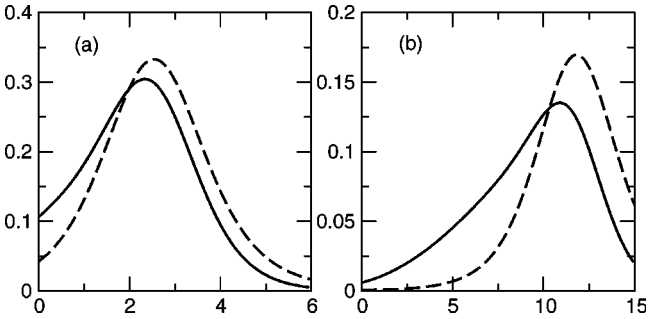

FIG. 7. Electron energy spectra predicted by TDSE (full line) and CV calculations (dashed line) for $\tau = 5$ a.u. (a) and 10 a.u. (b). The laser field amplitude is $F_o = 1$ a.u. and the photon energy is $\omega = 0.05$ a.u.

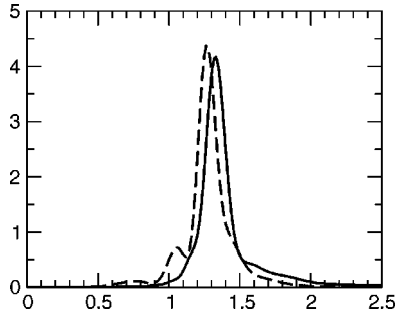

FIG. 8. Electron energy spectra predicted by TDSE (full line) and CV calculations (dashed line) with Ti:sapphire laser features achieved by Wien (see text): $I_0=3.5\times10^{14}$ W/cm^2 and $\tau=4$ fs.

trum. The field essentially exhibits a classical aspect, and the electron absorbs the corresponding classical energy, as defined above. Then the CV theory will be the most adapted tool to investigate atom ionization when laser sources with higher intensities and shorter pulse durations become available.

D. Total ionization probability

Within the range where it applies, the CV approach reproduces very well the energy and angular distributions. Then the question is how it compares with well-known previous theories in predicting the total ionization probability. Again the question is addressed here using TDSE results as a point of reference. In the community of experimentalists, the most popular theories are the approaches of ADK, Keldysh, and Landau (see references in Ref. [11]). However, no comparison can be made with the ADK model, because the total ionization probability is a time average over one period of an oscillating field with a constant amplitude. Thus the effect of a quickly changing amplitude cannot be reproduced. In view of remarks made by Bauer and Mulser in a recent study [11], it appears more convenient to compare the total ionization probability obtained by integrating both Keldysh and Landau ionization rates over the pulse duration after the constant electric field is replaced by the time dependent laser pulse field. CV and TDSE total ionization probabilities are obtained by a straightforward integration of energy distributions. Results for the ionization of H(1s) by pulses of increasing peak field amplitudes, but of the same duration and of the same frequency, are shown in Fig. 9. It is clear that CV predictions are very good whatever the intensity of the laser field, thus indicating that the CV approach works even for very weak fields, i.e., in perturbative conditions. Conversely, Landau's predictions are very poor, except for strong enough fields, where all theories predict a probability of ionization equal to 1 (saturation regime). Further, Landau's theory predicts saturation well before it occurs. Keldysh's theory agrees better with exact TDSE results for field amplitudes above 0.3 a.u., but also fails to predict the ionization probability at lower laser intensities.

Now it is worth noting that in the saturation regime, the relevant information is given by energy and angular distributions. The latter cannot be known either from Keldysh's or from Landau's theories. This feature strengthens the importance of a specific approach when laser-atom interactions occur far away from adiabatic conditions. For short enough pulses, our CV approach appears well adapted to such a situation.

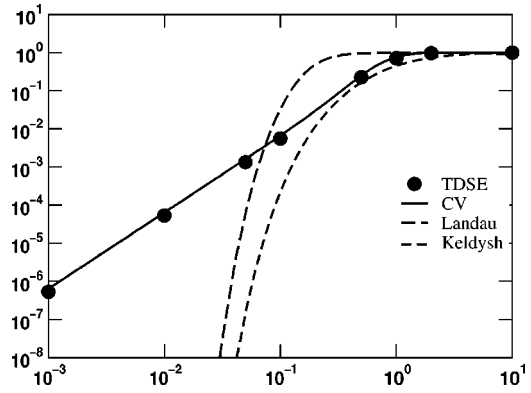

FIG. 9. Total ionization probability of a H(1s) atom as a function of the laser field amplitude. The laser pulse duration is $\tau=3$ a.u., and the photon energy is 0.05 a.u.

IV. CONCLUSION

We have shown that the well-known Coulomb-Volkov state is an accurate approximation of the solution of the time-dependent Schrödinger equation to investigate the ionization of hydrogen targets by intense and short enough laser pulses. The projection of this state onto a final continuum Coulomb wave function can provide accurate energy and angular distributions of the ejected electrons. Whatever the laser field amplitude, i.e., both in perturbative and nonperturbative regimes, this method gives reliable predictions as long as the sudden approximation is true, more precisely when $\tau \leqslant T_{n,Z}/2$, where $T_{n,Z}$ is the initial orbital period of the ejected electron. In addition, the electric field must not perform more than two oscillations during the pulse. Now, although the CV theory takes into account the influence of the Coulomb field of the nucleus in the structure of the unperturbed initial and final atomic states, this influence on the dynamics of ionization is ignored during the laser-atom interaction. As a result, the CV theory predicts electron spectra which are slightly shifted toward high energies. This feature is confirmed by ''exact'' numerical calculations. Therefore, CV predictions are accurate as long as the influence of the nuclear field on the dynamics of the process may be neglected, i.e., when laser pulses are either very short or very intense.

One advantage of our method is that it can be easily applied to more complicated systems than the hydrogen atom. It is especially true when two active electrons are considered. Indeed, ionization predictions with ''exact'' numerical calculations for two or more active electron systems appear very involved. Conversely, CV calculations may be easily performed with suitable correlated wave functions. Calculations for alkaline and helium targets are underway.

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APPENDIX A

The aim of this section is to introduce a bound CV state. We showed in Sec. III C how we can obtain the continuum CV wave function. We now show how an analytical expression of a bound CV state may be obtained from the continuum one. The electron energy distribution is obtained from $|\langle\chi_f^-|\varphi_i\rangle|^2$ with an integration over the ejection angles when the laser-atom interaction is completed. A further integration over the energy provides the total ionization probability. For a given energy,

$$|T_{fi}|^2=\lim_{t\to 0}|\langle f_0\mathcal{L}^-|\varphi_i\rangle|^2$$
$$=\lim_{t\to 0}\left|\int d\vec{r}\varphi_f^{-*}\exp\left[-i\vec{A}^-(t)\cdot\vec{r}+\frac{i}{2}\int_\tau^t dt' A^{-2}(t')\right.\right.$$
$$\left.\left.+i\vec{k}\cdot\int_\tau^t dt'\vec{A}^-(t')\right]\varphi_i\right|^2, \tag{A1}$$

where φ_f^- is the continuum Coulomb wave function. Now we are looking for an alternative form of the transition amplitude [Eq. (A1)]. First let us consider both vector potentials for an outgoing CV state and for an ingoing one:

$$\vec{A}^+(t)=-\int_0^t dt'\vec{F}(t'), \tag{A2}$$

$$\vec{A}^-(t)=-\int_\tau^t dt'\vec{F}(t'). \tag{A3}$$

From Eqs. (A2) and (A3), it is clear that

$$\vec{A}^-(0)=-\vec{A}^+(\tau), \tag{A4}$$

and it is easily shown that

$$\vec{A}^+(t)-\vec{A}^-(t)=-\int_0^\tau dt\vec{F}(t)=\vec{A}_0. \tag{A5}$$

Let us examine the term $(i/2)\int_\tau^0 dtA^{-2}(t)$. According to Eq. (A5), it may be written as

$$\frac{i}{2}\int_\tau^0 dtA^{-2}(t)=-\frac{i}{2}\left[\int_0^\tau dtA^{+2}(t)-\int_0^\tau dt2\vec{A}_0\cdot\vec{A}^+(t)\right.$$
$$\left.+\int_0^\tau dtA_0^2\right]. \tag{A6}$$

Then, from the above expressions, the exponential factor in Eq. (A1) becomes

$$\lim_{t\to 0}\exp\left(-i\vec{A}^-(t)\cdot\vec{r}+\frac{i}{2}\int_\tau^t dt' A^{-2}(t')\right)$$
$$=\lim_{t\to\tau}\exp\left(i\vec{A}^+(t)\cdot\vec{r}-\frac{i}{2}\int_0^t dt' A^{+2}(t')\right), \tag{A7}$$

where the constant phase factor

$$\exp\left(i\vec{k}\cdot\int_\tau^0 dt\vec{A}^-(t)+\frac{i}{2}\int_0^\tau dt2\vec{A}_0\cdot\vec{A}^+(t)-\frac{i}{2}\int_0^\tau dtA_0^2\right) \tag{A8}$$

is deliberately omitted since any constant phase factor may be ignored in the transition amplitude. We keep the term $\int_0^t dt' A^{+2}(t')$ to have a CV bound state which tends to a Volkov state at very high intensities. Thus $|T_{fi}|$ can be rewritten as

$$|T_{fi}|^2=\lim_{t\to\tau}\left|\int d\vec{r}\varphi_f^{-*}\exp\left(i\vec{A}^+(t)\cdot\vec{r}-\frac{i}{2}\right.\right.$$
$$\left.\left.\times\int_0^t dt' A^{+2}(t')\right)\varphi_i\right|^2. \tag{A9}$$

Now, from the quantum theory, we can exactly write

$$|T_{fi}|^2=\lim_{t\to 0}|\langle\Psi_f^-|\varphi_i\rangle|^2=\lim_{t\to\tau}|\langle\varphi_f^-|\Psi_i^+\rangle|^2, \tag{A10}$$

where Ψ_f^- and Ψ_i^+ are the exact solutions of the time-dependent Schrödinger equation for both a continuum state and a bound state, respectively. Since Ψ_f^- can be approximated by a continuum CV wave function χ_f^-, and keeping in mind the analogy between the transition amplitudes (A1) and (A9), Ψ_i^+ may be approximated in Eq. (A10) by an outgoing bound CV state χ_i^+ defined as

$$\chi_i^+(\vec{r},t)=\varphi_i(\vec{r},t)\exp\left(i\vec{A}^+(t)\cdot\vec{r}-\frac{i}{2}\int_0^t dt' A^{+2}(t')\right). \tag{A11}$$

APPENDIX B

Let us now evaluate the relative error induced by replacing $f^-(\vec{r},t)$ by $f_0(\vec{r})$. Most contributions to the transition amplitude come from a region where $\varphi_i(\vec{r},t)$ is nonnegligible. It is clear that the transition from a bound state to a continuum state occurs in this region. Thus it appears reasonable to average each function or operator O in this region on the initial state as follows: $\langle O\rangle=\langle\varphi_i|O|\varphi_i\rangle$. Under these considerations, Eq. (2.18) is now

$$\langle f^-(\vec{r},t)\rangle=\langle f_0(\vec{r})\rangle-\left\langle\int_\tau^t dt'\vec{A}(t')\cdot\vec{\nabla}f^-(\vec{r},t')\right\rangle, \tag{B1}$$

where $\langle\int_0^t dt' A(t')(\partial f^-(\vec{r},t')/\partial z)\rangle$ is the absolute error on $f^-(\vec{r},t')$. Then we can define a relative error as

$$\mathfrak{E}(t)=\left|\frac{\left\langle \int_0^t dt' A(t')\frac{\partial f^-(\vec{r},t')}{\partial z}\right\rangle}{\langle f^-(\vec{r},t')\rangle}\right|. \tag{B2}$$

If $f_0(\vec{r})$ is a good approximation of $f^-(\vec{r},t)$, we can substitute f_0 for f^- in Eq. (B2) assuming that $\mathfrak{E}\ll 1$. Since $\mathfrak{E}(t)$ depends on time, a relevant self-consistent criterion of accuracy has to be defined after the laser-atom interaction is completed. Taking $\mathfrak{E}(t)$ at time τ is not representative of the physical process because the electron can be ejected at any time. Then, it is better to average the error over the pulse duration. Finally, our criterion is given by

$$\mathfrak{E}=\frac{1}{\tau}\int_0^\tau dt\left|\int_0^t dt' A(t')\right|\times\left|\frac{\langle \partial f_0/\partial z\rangle}{\langle f_0\rangle}\right|. \tag{B3}$$

Theoretically, $\mathfrak{E}\ll 1$ implies f_0 to be a good approximation of f^-. Now we can simplify Eq. (B3) in the case of a very short laser pulse. In our calculations, we have generally $\omega \ll 2\pi/\tau$ so that the electric field reads

$$\vec{F}(t)=\begin{cases}\vec{F}_0\sin^2\left(\dfrac{\pi t}{\tau}\right) & \text{if } 0<t<\tau\\ \vec{0} & \text{elsewhere.}\end{cases} \tag{B4}$$

Under this condition, we obtain $A(t)=-\int_0^t dt' F(t')\approx -F_0 t/2$, and Eq. (B3) becomes

$$\mathfrak{E}=\frac{F_0\tau^2}{12}\left|\frac{\langle \partial f_0/\partial z\rangle}{\langle f_0\rangle}\right|=\beta(k)\frac{F_0\tau^2}{12}. \tag{B5}$$

It is noteworthy that β depends only on the energy of the electron for a given bound state. Since the energy distribution of the ejected electrons is close to the classical energy for very intense laser pulses, it is interesting to know the error for this particular value of the energy. In this case, the electron energy E_k is a function of the field parameters:

$$E_k\approx\frac{A^2(\tau)}{2}=\frac{F_0^2\tau^2}{8}. \tag{B6}$$

Finally, taking into account Eq. (B6) and $E_k=k^2/2$, we rewrite Eq. (B5) as

$$\mathfrak{E}=k\beta(k)\times\frac{\tau}{6}. \tag{B7}$$

$\beta(k)$ is a complicated expression, which is evaluated numerically. Omitting the normalization factor, f_0 is given by

$$f_0(\vec{r})\sim F_1(-i\nu,1,-ikr-i\vec{k}\cdot\vec{r})\sim F_1(-i\nu,1,-ikr-ik_z z). \tag{B8}$$

Now, for very intense fields, one may assume $k\simeq k_z$, because the electron is principally ejected along the field polarization. Finally, we have to evaluate numerically $\beta(k)$ defined by

$$\beta(k)=\frac{\left|\int d\vec{r}\,\varphi_i^*\dfrac{\partial f_0}{\partial z}\varphi_i\right|}{\left|\int d\vec{r}\,\varphi_i^* f_0\varphi_i\right|}. \tag{B9}$$

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