Ionization of hydrogen targets by short laser pulses

P. A. Macri, J. E. Miraglia, and M. S. Gravielle

Instituto de Astronomía y Física del Espacio, Consejo Nacional de Investigaciones Científicas y Técnicas, Casilla de Correo 67, Sucursal 28, 1428 Buenos Aires, Argentina

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We present a distorted-wave formulation of atomic ionization by short laser pulses based on Coulomb–Volkov states. The method is applied to atomic-hydrogen targets, for different interaction times and frequencies. Results are compared with the predictions of an exact numerical treatment, and good agreement is obtained as long as the convergence conditions of the perturbative series hold. The applicability of the method depends on the field intensity but not on the pulse duration, permitting a unified description of various ionization mechanisms.

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1. INTRODUCTION

Nowadays laser facilities are applied to a wide range of systems. In particular, intense and short laser pulses permit the generation of highly ionizated plasmas, which become useful sources of short-lived x rays with which to study chemical, biological, and material processes.[1]

The power of exact numerical methods for computing the solution of the time-dependent Schrödinger equation seems to explain all the physics behind the ionization of hydrogen by short laser pulses.[2–4] However, when intensities are high and the duration of the pulse long, as well as for many-electron atoms, these methods become impracticable. For this reason we investigate a more analytical approach to the solution based on the well-known Coulomb–Volkov (CV) wave functions. In previous studies[5,6] these approximate solutions of the time-dependent problem were applied in the framework of sudden approximation. Accurate results were obtained in comparison with exact computations, as long as the duration of the pulse did not exceed half of the initial states orbital period and the laser field did not complete more than two optical cycles. At present, laser facilities have reached femtosecond and subfemtosecond pulses,[7] and thus the sudden Coulomb–Volkov (SCV) approximation gives reliable ionization results for large principal quantum numbers, i.e., for $n \geqslant 4$.[6] However, for application to hydrogen targets in the ground state, the SCV approximation requires pulse durations shorter than 8 attoseconds, which are still far from being experimentally obtained. Therefore a simple and reliable approach to predicting electron distributions for longer pulses and for atoms in their lower quantum states is needed.

After their proposal in Ref. 8, CV functions have been extensively used to compute processes driven by monochromatic, low-frequency, and low-intensity lasers.[9–12] In the research reported here, these functions are employed within the framework of a distorted wave formalism. The method is applied to ionization of atomic hydrogen in its ground state by a strong laser field without any restrictions on pulse duration or on the number of oscillations that the laser pulse can perform. The paper is organized as follows: in Section 2 we derive generalized CV wave functions. In Section 3 we present the time-dependent distorted-wave theory. Results are presented in Section 4, and they are compared with exact calculations and with the previous SCV values. Finally, in Section 5 we discuss our main conclusions.

Atomic units (a.u.) are used throughout unless otherwise stated.

2. THEORY

A. Description of the Laser Pulse

We consider the interaction of a target atom with an ultrashort laser pulse described by a time-dependent electric field linearly polarized along the z axis:

$$\mathbf{F}(t) = \begin{cases} \mathbf{F}_0 \sin(\omega t + \varphi)\sin^2(\pi t/\tau) & 0 < t < \tau \\ \mathbf{0} & \text{elsewhere} \end{cases} \tag{1}$$

where τ is the duration of the envelope, ω is the photon energy, and phase φ is selected as $\varphi = -\omega\tau/2 + \pi/2$ for a symmetric pulse. $\mathbf{F}(t)$ is related to vector potential $\mathbf{A}^{\pm}(t)$ by

$$\mathbf{A}^{\pm}(t) = -\int_{\mp\infty}^{t} \mathrm{d}t' \mathbf{F}(t'), \tag{2}$$

where we have set $\mathbf{A}^{\pm}(\mp\infty) = \mathbf{0}$. For ultrashort pulses the electric field does not perform oscillations, and its net integral is always different from zero. In this case the ionization mechanism is in a collisional regime,[5] so called because of the similarities between the effect of the electromagnetic pulse and that produced by the impact of a fast ion. For longer pulse durations, however, the laser frequency tends to the photon energy, and electron production is the result of other mechanisms. For weak fields leading to small probabilities, multiphoton ionization is found. For strong fields there is a critical strength $E_0 = Z^3/4n^2$ at which the bound electrons begin to move

out freely above the resultant potential barrier, leading to over-the-barrier ionization. Just below that critical field strength, tunnel ionization is the proper mechanism.

B. Distorted-Wave Functions

In the presence of an external electric field the evolution of electronic state $\Psi(t)$ is determined by the time-dependent Schrödinger equation

$$i\frac{\partial\Psi(\mathbf{r},t)}{\partial t} = [H_0 + V(t)]\Psi(\mathbf{r},t), \tag{3}$$

where $H_0 = -\nabla^2/2 - Z/r$ is the atomic Hamiltonian and $V(t) = \mathbf{r}\cdot\mathbf{F}(t)$ is the electric field perturbation in the length gauge. As a consequence of this interaction, one electron initially bound to the target nucleus in state ϕ_i is emitted in a continuum state ϕ_f with momentum $\mathbf{k}_f$. Here we introduce a simple approximation for the wave function corresponding to an electron in the simultaneous presence of a Coulomb charge Z and a plane-wave electromagnetic field and use the latter within a distorted wave formalism. For $Z = 0$, an exact solution of Eq. (3) is given by the Volkov state[13,14]:

$$\Psi_{\mathbf{k}}{}^{V}(\mathbf{r},t) = \exp(-i\varepsilon t)\exp(i\mathbf{k}\cdot\mathbf{r})\exp[iD^{-}(\mathbf{k},\mathbf{r},)], \tag{4}$$

where

$$D^{\pm}(\mathbf{k},\mathbf{r},t) = \mathbf{A}^{\pm}(t)\mathbf{r} - \mathbf{k}\int_{\mp\infty}^{t} t'\mathbf{A}^{\pm}(t') - {}^1\!/_2\int_{\mp\infty}^{t} t'[\mathbf{A}^{\pm}(t')]^2, \tag{5}$$

and ε is the electron energy.

For $Z \neq 0$ we can derive an approximate solution of time-dependent Schrödinger equation (3) by using the well-known impulse approach.[15,16] We propose a state that we call here an impulse Coulomb–Volkov (ICV) state as the initial distorted wave function:

$$\chi_i{}^{ICV+}(\mathbf{r},t) = \frac{1}{(2\pi)^{3/2}}\int d\mathbf{k}\exp(-i\varepsilon_i t)\tilde{\varphi}_i(\mathbf{k}) \times \exp[i\mathbf{k}\cdot\mathbf{r} + iD^{+}(\mathbf{k},\mathbf{r},t)], \tag{6}$$

$$= \phi_{\mathbf{i}}[\mathbf{r} - \alpha^{+}(t)]\exp[iD^{+}(\mathbf{0},\mathbf{r},t)], \tag{7}$$

where $\phi_i(\mathbf{r},t) = \exp(-i\varepsilon_i t)\varphi_i(\mathbf{r})$ is the initial bound state in the absence of the field and a tilde denotes Fourier transformation in momentum space. Vector $\alpha^{+}(t) = \int_{-\infty}^{t} dt'\mathbf{A}^{+}(t')$, which satisfies $\ddot{\alpha}^{+}(t) = -\mathbf{F}(t)$, represents the classic displacement of the free electron from its center of oscillation in a radiation field $\mathbf{F}(t)$. Therefore the ICV state given by Eq. (7) is obtained from the unperturbed state φ_i that replaces the plane wave associated with the initial electronic momentum distribution with a field-dependent phase of the Volkov state. This is the same procedure as the one usually employed for heavy-particle collisions.[16,17] We can also verify that wave function $\chi_i{}^{\mathrm{ICV+}}$ tends to unperturbed wave function ϕ_i as $t \to -\infty$, satisfying the correct asymptotic conditions.

The use of $\chi_i{}^{\mathrm{ICV+}}$ in the computation of ionization probabilities leads us to two-center Coulomb integrals, and we intend to evaluate them in future research. We obtain simpler approximation of the initial ICV state by neglecting the term $\alpha^{+}(t)$ in the argument of ϕ_i, which leads to an initial CV wave function. This approximation is particularly valid for large frequencies for which quiver amplitude $\alpha_0 = F_0/\omega^2$ is small compared with the average electron position in the nucleus field and for ultrashort pulses for which there is not enough time for the electron to reach α_0. However, CV states have been used to describe a variety of processes, regardless of specific laser parameters. In the same spirit, we apply CV states to describe photoelectron production and then to analyze its usefulness. The initial CV distorted wave function reads as

$$\chi_i{}^{\mathrm{CV+}}(\mathbf{r},t) = \phi_i(\mathbf{r},t)\exp[iD^{+}(0,\mathbf{r},t)]. \tag{8}$$

In similar way, we can derive a final ICV wave function, from which we can obtain the previously deduced[8] final CV state:

$$\chi_f{}^{\mathrm{CV-}}(\mathbf{r},t) = \phi_f(\mathbf{r},t)\exp[iD^{-}(\mathbf{k}_f,\mathbf{r},t)], \tag{9}$$

where $\phi_f(\mathbf{r},t) = \varphi_f{}^{-}(\mathbf{r})\exp(-i\varepsilon_f t)$ is the final Coulomb wave function with momentum $\mathbf{k}_f$.

C. Time-Dependent Distorted-Wave Theory

In this section we describe the application of CV states given by Eqs. (8) and (9) as distorted functions that take into account part of perturbation $V(t)$. These distorted wave functions satisfy the asymptotic conditions $\chi_i{}^{\mathrm{CV+}}(t) \to \phi_i(t)$ for $t \to -\infty$ and $\chi_f{}^{\mathrm{CV-}}(t) \to \phi_f(t)$ for $t \to +\infty$. The probability of transition $\phi_i \to \phi_f$ can be expressed as $P_{fi} = |T_{fi}{}^{\pm}|^2$, where $T_{fi}{}^{+(-)}$ is the *post* (*prior*) form of the transition amplitude, which reads as

$$T_{fi}{}^{+} = \lim_{t\to+\infty}\langle\phi_f(t)|\Psi_i{}^{+}(t)\rangle \quad \textit{post} \text{ form}, \tag{10}$$

$$T_{fi}{}^{-} = \lim_{t\to-\infty}\langle\Psi_f{}^{-}(t)|\phi_i(t)\rangle \quad \textit{prior} \text{ form}, \tag{11}$$

where $\Psi_i{}^{+}(t)[\Psi_f{}^{-}(t)]$ is the exact electronic wave function with outgoing (incoming) asymptotic conditions; i.e., $\Psi_i{}^{+}(t) \to \phi_i(t)$ as $t \to -\infty$ $[\Psi_f{}^{-}(t) \to \phi_f(t)$ as $t \to +\infty]$.

Within this formalism, Eqs. (10) and (11) can be rewritten in terms of distorted CV functions (8) and (9) as

$$T_{fi}{}^{+} = a_{fi}{}^{-} - i\int_{-\infty}^{+\infty} dt\langle\chi_f{}^{\mathrm{CV-}}(t)|W_f{}^{\dagger}(t)|\Psi_i{}^{+}(t)\rangle, \tag{12}$$

$$T_{fi}{}^{-} = a_{fi}{}^{+} - i\int_{-\infty}^{+\infty} dt\langle\Psi_f{}^{-}(t)|W_i(t)|\chi_i{}^{\mathrm{CV+}}(t)\rangle, \tag{13}$$

with

$$a_{fi}{}^{+} = \lim_{t\to+\infty}\langle\phi_f(t)|\chi_i{}^{\mathrm{CV+}}(t)\rangle \quad \textit{post} \text{ form}, \tag{14}$$

$$a_{fi}{}^{-} = \lim_{t\to-\infty}\langle\chi_f{}^{\mathrm{CV-}}(t)|\phi_i(t)\rangle \quad \textit{prior} \text{ form}. \tag{15}$$

The potentials W_j are the corresponding distortion potentials, defined by

$$\left(H(t) - i\frac{\mathrm{d}}{\mathrm{d}t}\right)|\chi_j^{\mathrm{CV}s}(t)\rangle = W_j(t)|\chi_j^{\mathrm{CV}s}(t)\rangle, \quad (16)$$

where $j = i, f$ and s is + and −, respectively. More specifically, we can write

$$W_i(t)\chi_i^{\mathrm{CV}+}(\mathbf{r}, t) = -\iota\nabla_{\mathbf{r}}\phi_i(\mathbf{r}, t) \cdot \mathbf{A}(t)\exp[iD^{+}(\mathbf{0}, \mathbf{r}, t)], \quad (17)$$

$$W_f(t)\chi_f^{\mathrm{CV}-}(\mathbf{r}, t) = -\iota[\nabla_{\mathbf{r}}\phi_f(\mathbf{r}, t) - \iota\mathbf{k}_f] \cdot \mathbf{A}(t)\exp[iD^{+-} - (\mathbf{k}_f, \mathbf{r}, t)]. \quad (18)$$

It is worth noting that Eqs. (12) and (13) are nothing but time-dependent equivalents of the well-known two-potential formulas.[18] An alternative presentation of the time-dependent formalism can be found in Ref. 19.

Calculation of the exact transition amplitude involves knowledge of the electronic wave function $\Psi_i^{+}(t)$ or $\Psi_f^{-}(t)$, which, until now, could be achieved only through numerical computation. However, from Eqs. (12) and (13) we can derive different approximations, replacing the exact wave function with unperturbed (singly distorted, SD) or distorted (doubly distorted, DD) approximations. For the *post* form we obtain

$$T_{fi}^{\mathrm{SD}+} = a_{fi}^{-} - i\int_{-\infty}^{+\infty} t\langle\chi_f^{\mathrm{CV}-}(t)|W_f(t)|\phi_i(t)\rangle = -i\int_{-\infty}^{+\infty} t\langle\chi_f^{\mathrm{CV}-}(t)|V(t)|\phi_i(t)\rangle, \quad (19)$$

$$T_{fi}^{\mathrm{DD}+} = a_{fi}^{-} - i\int_{-\infty}^{+\infty} t\langle\chi_f^{\mathrm{CV}-}(t)|W_f(t)|\chi_i^{\mathrm{CV}+}(t)\rangle, \quad (20)$$

and, in the *prior* form,

$$T_{fi}^{\mathrm{SD}-} = a_{fi}^{+} - i\int_{-\infty}^{+\infty} t\langle\phi_f(t)|W_i(t)|\chi_i^{\mathrm{CV}+}(t)\rangle = -i\int_{-\infty}^{+\infty} t\langle\phi_f(t)|V(t)|\chi_i^{\mathrm{CV}+}(t)\rangle, \quad (21)$$

$$T_{fi}^{\mathrm{DD}-} = a_{fi}^{+} - i\int_{-\infty}^{+\infty} t\langle\chi_f^{\mathrm{CV}-}(t)|W_i(t)|\chi_i^{\mathrm{CV}+}(t)\rangle. \quad (22)$$

Notice that in the DD model the *post* and *prior* versions are equivalent, that is, $T_{fi}^{\mathrm{DD}+} = T_{fi}^{\mathrm{DD}-}$, whereas in the SD theory a *prior–post* discrepancy appears. The second form of Eq. (19) is just the Keldysh–Faisal–Reiss[20–22] or strong-field-approximation ionization rate, with CV functions replacing Volkov states.

The use of CV states in Eqs. (14) and (15), however, leads to the SCV transition amplitudes as defined in Ref. 5:

$$T_{fi}^{\mathrm{SCV}} = a_{fi}^{+} = a_{fi}^{-}$$

This means that the distorted-wave approach proposed here includes two separate terms. The first term is more relevant for ultrashort pulses, when ionization by a collisional mechanism is dominant. For many field oscillations this term tends to zero and is necessary for describing the process through the time integral over all intermediate transitions given by the second term.

3. RESULTS AND DISCUSSION

For a given energy, the angular distribution of the ejected electrons is

$$\frac{\partial P_{fi}}{\partial E_k \partial\Omega_k} = k|T_{fi}|^2, \quad (23)$$

whereas integration over the direction of the ejected electrons yields the energy distribution

$$\frac{\partial P_{fi}}{\partial E_k} = k\int \mathrm{d}\Omega_k|T_{fi}|^2. \quad (24)$$

To evaluate the transition amplitude we employ the DD model [Eq. (20) or (22)] and the *post* and *prior* forms of the SD models [Eqs. (19) and (21)]. By employing the CV functions we can evaluate the matrix elements inside the SD and DD transition amplitudes as closed analytical forms,[23] evaluating numerically the remaining integrals in time and angle. Results are compared with the SCV approximation[5,6] and with the exact calculation of the time-dependent Schrödinger equation.[2]

Our goal is to propose a reliable theory, regardless of the duration of the pulse, τ. Here we present calculations with values of τ from subfemtoseconds to femtoseconds. The field strength is $F_0 = 0.1$ a.u., corresponding to the range of strong fields but still in the perturbative regime. The method is not valid for higher values of F, for which ionization is produced in an effective time much shorter than the pulse duration itself. However, we vary the ω value to permit no to many oscillations of the field

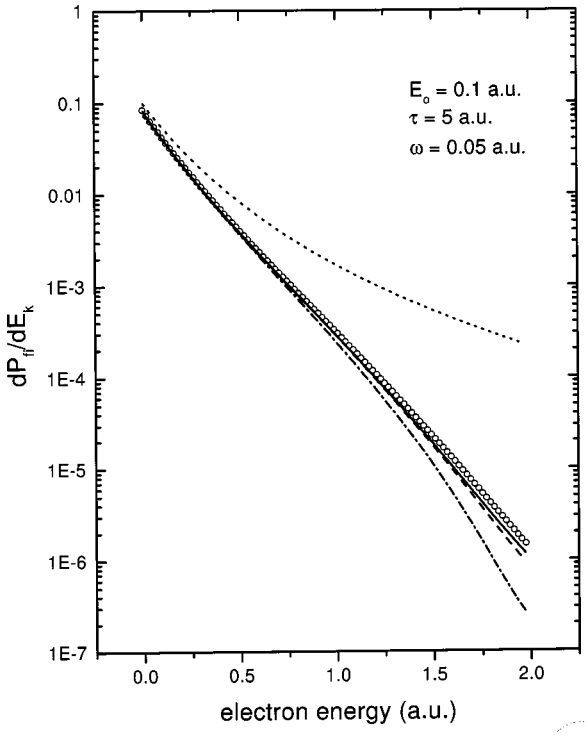

Fig. 1. Electron energy spectra of hydrogen ionization predicted by the time-dependent Schrödinger equation (open circles), (SD)+ (solid curve), (SD)− (dotted–dashed curve), and (DD) (dashed curve) models and by the SCV approach (dotted curve) for laser parameters $E_0 = 0.1$ a.u., $\tau = 5$ a.u., and $\omega = 0.05$ a.u.

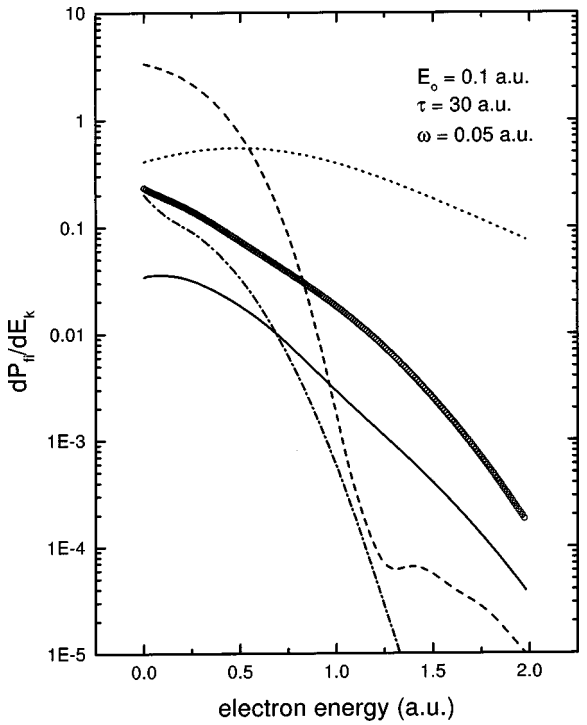

Fig. 2. Same as Fig. 1 but for laser parameters $E_0 = 0.1$ a.u., $\tau = 30$ a.u., and $\omega = 0.05$ a.u.

inside the envelope. The first case corresponds to the collisional mechanism, whereas the second case corresponds to an OBI process.

Let us consider first that the laser field does not oscillate and acts as an impinging fast ion (Figs. 1 and 2). In Fig. 1 we show the electron energy distribution for ionization by a laser pulse with parameters $\tau = 5$ a.u. and $\omega = 0.05$ a.u. We observe excellent agreement between the distorted wave approaches and the exact computation of the time-dependent problem. This agreement holds true even when the duration of the pulse is decreased below the half-orbital period, that is, in the domain of the sudden approximation. For large electron energies we find the best (worst) agreement when distortion in the final (initial) state is taken into account. SCV results, as expected, are not highly accurate for these laser parameters: They diverge from the exact calculations practically at threshold. In Fig. 2, results for a longer pulse, $\tau = 30$ a.u., with the same frequency, $\omega = 0.05$ a.u., are displayed. In this case we find qualitative agreement only between single distortion in the final state, SD+, and exact computation of the time-dependent Schrödinger equation. Although SD− theory gives the correct threshold behavior, it soon diverges from the correct results, whereas the DD theory fails in the whole energy range. Furthermore, we can see that the SCV approximation presents a maximum, absent from the exact calculations, at an energy $E_C + \epsilon_i \sim 0.5$ a.u., where

$$E_C = 1/2 \left| \int_0^\tau dt F(t) \right| \tag{25}$$

is the classic transferred energy. For the cases considered above, the pulse duration ($\tau = 5$ a.u. in Fig. 1 and $\tau = 30$ a.u. in Fig. 2) is short enough to prevent oscillations. Having considered this and other cases with similar results, we conclude that present distorted-wave theories are better for the lowest transferred energies E_C. This result is in agreement with the concept that small potentials lead to faster convergence of the perturbative series.

We now test the opposite case, i.e., that when the laser field performs many oscillations inside the pulse envelope (Figs. 3 and 4). In Fig. 3 we show the electron energy distribution for ionization by a laser with parameters $\tau = 30$ a.u. and $\omega = 1$ a.u.. We observe excellent agreement between the distorted-wave approach and the exact calculations, whereas the SCV approximation does not display any structure in the spectrum. This is so because in the SCV approximation the transition amplitude is given by overlapped Eqs. (14) and (15), and intermediate states are not taken into account. As in Fig. 1, we obtain the best results by distorting the final state. We have found perfect agreement for single distortion in the final state, SD+, and minor discrepancies with the DD approximation. For single distortion in initial state SD− we still find reliable results at moderate electron energies.

Results for a longer pulse, $\tau = 150$ a.u., with a lower frequency of $\omega = 0.25$ a.u., are displayed in Fig. 4. We observe that, again, the best agreement is obtained for single distortion in final state SD+, although a shift of the maxima to large energies and a scale difference between the distorted-wave theory and the exact results are present here. We argue that the dressing of the initial state in this case introduces spurious ponderomotive Stark shifts of the above-threshold ionization peaks in the SD− calculations (in state DD this spurious shift is partially balanced by the more-accurate shift incorporated in the final state).

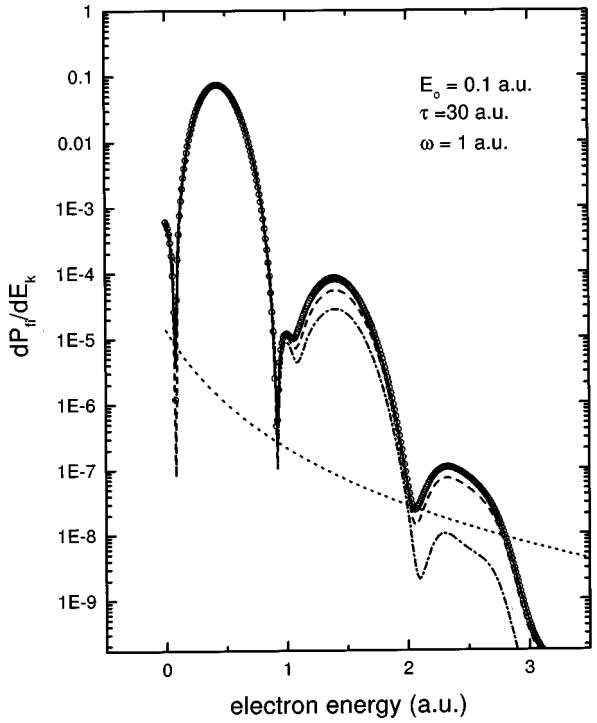

Fig. 3. Same as Fig. 1 but for laser parameters $E_0 = 0.1$ a.u., $\tau = 30$ a.u., and $\omega = 1$ a.u.

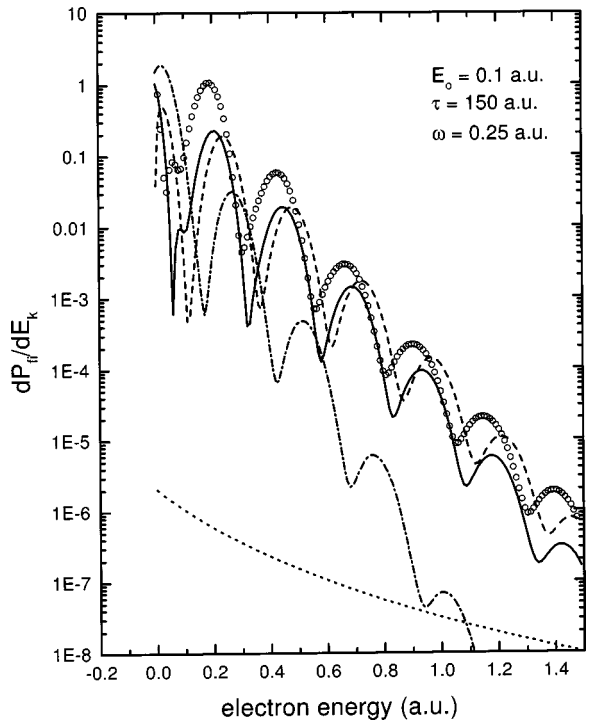

Fig. 4. Same as Fig. 1 but for laser parameters $E_0 = 0.1$ a.u., $\tau = 150$ a.u., and $\omega = 0.25$ a.u.

For Figs. 3 and 4 we considered laser parameters that let the electric field perform many (approximately five) oscillations and thus for which the laser frequency tends to the photon energy. At this point we can distinguish two regimes according the value $\gamma = \omega/\epsilon_i$, the ratio of the photon and bound state energies. The condition that $\gamma > 1$, satisfied by the system parameters of Fig. 3, implies that the behavior of the electrons is dominated by the laser rather than by the atomic field. CV states are constructed precisely with this prescription because, as stated in Ref. 6, the electron dynamics during the interaction time are determined only by the external electric field. This explains the detailed and precise description of the process that we observe from Fig. 3. In Fig. 4, however, we have $\gamma < 1$, and in this situation the Coulomb potential affects the electron dynamics more than the laser pulse does; this make the CV functions less realistic states, as we can confirm by examining Fig. 4.

Differences among various distorted-wave approximations can be understood if we consider that the former assumption is more reliable for an electron moving in the continuum than for a bound state close to the nucleus. In any case, energy distributions are exactly the same for the absorption of the first photons. Because at low energies the first Born approximation works well (one photon absorbed with an energy higher than the binding energy), it is reasonable to expect similar results for any improved theory, such as the SD^+, SD^-, and DD theories.

4. CONCLUSIONS

In this paper, we have shown that CV states can be employed in the framework of distorted-wave theory to give an accurate description of hydrogen ionization by short laser pulses. Regardless of the duration of the pulse, this method gives reliable predictions as long as the necessary conditions for convergence of the perturbative series hold. In this way it is possible to compute with the same method collisional ionization and above-threshold ionization. Because CV states do not take into account the presence of the nucleus during the interaction time, single distortion in the final SD^+ state gives more reliable results. For above-threshold ionization processes, accuracy increases for photon energies greater than the initial bound energy.

This computationally inexpensive method is particularly useful for long durations of the pulse, for which exact methods present severe convergence difficulties, and for multielectronic species, for which numerical methods are impracticable.

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