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A simple non-perturbative approach of atom ionisation by intense and ultra-short laser pulses

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Abstract. A simple theoretical approach based on Coulomb-Volkov states is introduced to predict ionisation of atoms by intense laser pulses in cases where the effective interaction time does not exceed one or two optical cycles [M. Nisoli *et al.*, Opt. Lett. **22**, 522 (1997)]. Under these conditions, the energy distributions of ejected electrons predicted by this non-perturbative approach are in very good agreement with "exact" results obtained by a full numerical treatment. The agreement is all the better that the principal quantum number of the initial state is high. For very strong fields, most electrons are ejected at an energy E_C which is close to the classical kinetic energy that would be transferred to free electrons by the electromagnetic field during the pulse. The power of the present approach appears when $E_C > 1$ keV. In this region, full numerical treatments become very lengthy and finally do not converge. However, the present Coulomb-Volkov theory still makes reliable predictions in very short computer times.

PACS. 32.80.-t Photon interactions with atoms – 32.80.Fb Photoionization of atoms and ions

1 Introduction

The recent development of new laser facilities, which produce ultra-short and ultra-intense laser pulses, opens the way to new applications of laser-atom interactions. Indeed, pulses lasting for a few femtoseconds with high peak intensities (10^{17} Wcm^{-2} or higher) can now be generated. At the University Bordeaux 1, the laser facility CELIA produces short energetic pulses ($\lambda \approx 800$ nm, 20 fs, 20 mJ) at a frequency of 1 kHz and intensities $\sim 10^{18}$ Wcm^{-2} are within reach [1]. Focusing such pulses on small targets (aggregates, droplets, etc.) permits to create highly ionized plasmas, which are bright and short-lived X-ray sources of very small size [2]. These X-ray sources are used in chemistry or in biology to follow the time evolution of molecular processes. They might also be very useful in material sciences to detect tiny structures or small defects. Now, the evolution of the plasma, as well as its early growth, depend strongly on the ionisation processes [2]. In 1985, Lambropoulos [3] gave convincing arguments which show that "substantial ionisation during the rise of the pulse of a strong laser is inevitable". For peak power much above 10^{14} Wcm^{-2} at frequencies in the near infrared, Lambropoulos suggested that "it would be interesting to see what will happen as the pulse length approaches 10 fs or so". With the above mentioned parameters of the CELIA facility and assuming sine-square time envelope for the electric field of the pulse with a peak intensity of 10^{18} Wcm^{-2}, we performed a full numerical calculation which shows that a hydrogen atom H($1s$) is fully ionised in the first half cycle of the pulse, *i.e.* during less than 1.4×10^{-15} s. It is worth noting that the maximum laser field reached during this time is equivalent to a laser intensity of a few 10^{15} Wcm^{-2}. *A priori*, the study of processes induced by very intense laser fields, the maximum amplitude of which changes rapidly with time, calls for non-perturbative methods [4]. However, the full numerical treatment of non-perturbative theoretical approaches often requires long computer times, as well as wide memory [5,6]. Now, when the ionisation is completed in a very short time τ, thus defining an *effective pulse* that might be much smaller than the pulse duration itself (even well below 1 fs), it would be interesting to get the angular and energy distributions of the electrons at τ. Indeed, in dense plasmas, the subsequent propagation of an electron in the continuum may well be described by PIC codes [8]. Therefore, simpler approaches, that pertain to the "sudden approximation" in quantum mechanics [7], may be envisaged more especially in the case where τ is shorter than or comparable to the initial orbital period of an ejected electron [8].

Then, in the special case of ultra-intense laser pulses, it appears interesting to look for approximate non-perturbative approaches which should be much easier to implement than full numerical treatments and which could provide reliable data on ionisation *at the very beginning of the pulse*. Further, in case of multielectron transitions, the use of full numerical approaches [10] is made

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very difficult and often impossible because of the huge matrices to be handled while one may hope that approximate non-perturbative methods provide reliable predictions with reasonable computer times. Such a theory has already been introduced by Presnyakov [11]. Its application to short and intense pulses, which is investigated hereafter, is not fully satisfactory. However, Presnyakov's ideas guided us to introduce a new simple approach based on Coulomb-Volkov states [12]. The new method is presented here and the domain where it can provide accurate predictions on ionisation by short and intense laser pulses is marked out. The first Born approximation (FBA) is also investigated because it indicates the limits of the perturbative regime.

In this preliminary work, our study is concentrated on the ionisation of hydrogen atoms, both because predictions may be scaled to more complex orbitals and because it is easier to exhibit the physical meaning of results. In what follows, "exact" numerical predictions are compared with the present approach for effective pulses which are often much shorter than the present-day shortest ones.

The paper is organised as follows: theoretical approaches are presented in Section 2. Presnyakov's theory and the first Born approximation are briefly recalled in Section 2.1. The Coulomb-Volkov approach is sketched in Section 2.2. Results and discussion are given in Section 3. Conclusions are drawn in Section 4.

Atomic units are used throughout unless otherwise stated.

2 Theory

The evolution of the system is described by the time dependent Schrödinger equation which, in the length gauge and in the dipole approximation, has the form

$$\mathrm{i}\frac{\partial \Psi(\mathbf{r},t)}{\partial t} = \left(-\frac{\nabla^2}{2} - \frac{z}{r} + \mathbf{r}\cdot\mathbf{E}(t) \right) \Psi(\mathbf{r},t). \qquad (1)$$

Our study is made with a laser field linearly polarised along the z-axis. The finite duration of the pulse is featured through a sine-square envelope. Then, the field expression is:

$$\begin{cases} \mathbf{E}(t) = \mathbf{E}_0 \sin(\omega t + \varphi) \sin^2\left(\dfrac{\pi t}{\tau}\right) & \text{when} \quad t \in [0,\tau] \\ \mathbf{E}(t) = \mathbf{0} \quad \text{elsewhere.} \end{cases} \qquad (2)$$

In what follows, all calculations are made with a symmetric pulse, which implies $\varphi = \pi/2 - \omega\tau/2$. 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, except in a particular case that will be specified. To check the relevance of the approximate methods which are investigated in subsequent sections, we compared their predictions with "exact" numerical calculations based on a B-spline expansion of $\Psi(\mathbf{r},t)$ [5]. Hereafter, the latter are referred to as TDSE (Time Dependent Schrödinger Equation) calculations.

2.1 Presnyakov's theory and the first Born approximation

The theory developed by Presnyakov *et al.* [15] was first applied to study the double ionisation of helium by fast highly charged ions. Later on, Presnyakov [11] indicated that it might apply to the ionisation of hydrogen by intense laser pulses. In this approach, the perturbing field appears explicitly at each time in the electronic continuum wave functions. These later states are continuum Coulomb wave functions of the target with a time-dependent momentum $\mathbf{p}(t)$ which is

$$\mathbf{p}(t) = \mathbf{k} + \mathbf{A}(t) = \mathbf{k} - \int_0^t \mathrm{d}t' \mathbf{E}(t') \qquad (3)$$

where $\mathbf{k}$ is the final momentum of the ejected electron and $\mathbf{A}(t)$ is the vector potential defined by (3). Together with the discrete states of the target, these continuum states form a new orthogonal basis set which is complete at all times, thus leading to a set of new coupled equations for the transition amplitudes. If we consider that the initial population is not significantly depleted during the interaction, the electron removal probability may be written as

$$P_i = \int \mathrm{d}\mathbf{k} \left\{ 2\,\mathrm{Re} \int_0^\tau \mathrm{d}t \int_0^t \mathrm{d}t' U(t) U^*(t') \right\} \qquad (4)$$

where $U(t)$ is the dipole matrix element between the initial bound state and the continuum state of the basis at time t. Note that if we replace the continuum state by a standard stationary Coulomb state in Presnyakov's theory ($\mathbf{p}(t) = \mathbf{k}$), it turns into the first Born approximation (FBA), which permits to delimit the perturbative regime. The FBA transition amplitude is simply given by

$$T_{\mathrm{fi}} = -\mathrm{i} \int_0^\tau \mathrm{d}t \langle \phi_{\mathrm{f}} | \mathbf{r}\cdot\mathbf{E}(t) | \phi_{\mathrm{i}} \rangle \qquad (5)$$

where $\phi_{\mathrm{i}}(\mathbf{r},t)$ and $\phi_{\mathrm{f}}(\mathbf{r},t)$ are the initial and the final states respectively. Numerical calculations of the spectrum of ejected electrons which have been performed with the Presnyakov's theory are not satisfactory (see Sect. 3). Therefore, we seek for a new approximate theory, that can make reliable predictions with ultra-intense and ultra-fast laser pulses, *i.e.*, far outside the perturbation regime. This new approach based on Coulomb-Volkov (CV) wave functions [12] is introduced in the following section.

2.2 Coulomb-Volkov approach

We wish to predict the energy distribution of the ejected electrons after the interaction with the electromagnetic field. We start from the standard result of the quantum theory where the transition amplitude is given by

$$T_{\mathrm{fi}} = \lim_{t\to-\infty} \langle \Psi_{\mathrm{f}}^-(t) | \phi_{\mathrm{i}}(t) \rangle = \langle \Psi_{\mathrm{f}}^-(t) | \phi_{\mathrm{i}}(t) \rangle_{t=0} \qquad (6)$$

where $\phi_{\mathrm{i}}(\mathbf{r},t)$ is the initial bound state, and $\Psi_{\mathrm{f}}^-(\mathbf{r},t)$ is the exact solution of (1) identical to the final continuum

state $\phi_{\rm f}^{-}(\mathbf{r},t)$ at $t \to +\infty$. Since equation (1) does not admit an exact analytical solution, we replace $\Psi_{\rm f}^{-}(\mathbf{r},t)$ by an approximate CV wave function, which is explicitly:

$$\chi_{\rm f}^{-}(\mathbf{r},t) = \phi_{\rm f}^{-}(\mathbf{r},t)\exp\left\{\mathrm{i}\mathbf{A}^{-}(t)\cdot\mathbf{r} - \frac{\mathrm{i}}{2}\int_{\tau}^{t}\mathrm{d}t' A^{-^2}(t') - \mathrm{i}\mathbf{k}\cdot\int_{\tau}^{t}\mathrm{d}t'\mathbf{A}^{-}(t')\right\} = \phi_{\rm f}^{-}(\mathbf{r},t)L^{-}(\mathbf{r},t) \quad (7)$$

where $\phi_{\rm f}^{-}(\mathbf{r},t)$ is the final Coulomb wave function with momentum $\mathbf{k}$ and $\mathbf{A}^{-}(t) = -\int_{\tau}^{t}\mathrm{d}t'\mathbf{E}(t')$. The transition probability is $|T_{\rm fi}|^2$. It is easy to show that it may be changed into:

$$\left|T_{\rm fi}\right|^2 = \left|\left\langle\phi_{\rm f}^{-}(t)\middle|\chi_{\rm i}^{+}(t)\right\rangle_{t=\tau}\right|^2 \quad (8)$$

with

$$\chi_{\rm i}^{+}(\mathbf{r},t) = \phi_{\rm i}(\mathbf{r},t)\exp\left\{\mathrm{i}\mathbf{A}^{+}(t)\cdot\mathbf{r} - \frac{\mathrm{i}}{2}\int_{0}^{t}\mathrm{d}\tau\mathbf{A}^{+^2}(\tau)\right\} = \phi_{i}(\mathbf{r},t)L^{+}(\mathbf{r},t). \quad (9)$$

In (9), $\phi_{\rm i}(\mathbf{r},t)$ is the initial bound state, $\mathbf{A}^{+}(t) = -\int_{0}^{t}\mathrm{d}t'\mathbf{E}(t')$ and the exponential factor is a part of the phase which appears in standard Volkov states which are exact solutions for a free electron in an electric field. Indeed, standard Volkov states are not adapted to our case because they ignore the interaction of the electron with the nucleus. In our approach, this Coulomb interaction is taken into account by replacing, in Volkov states, the free plane wave-function by the initial bound wave-function. It is worth noting that the smaller the influence of the nuclear field on the variation of the electron energy during the pulse, the better the approximation. In addition, we have to assume that the field does not have time to oscillate. Otherwise, the vector potential as defined in (3) with the electric field given by (2) is all the closer to zero that the number of cycles within the square-sine envelope is large. Indeed, when $\mathbf{A}(\tau) \to \mathbf{0}$, the CV state (9) becomes an unperturbed bound state and the transition amplitude (8) vanishes. Therefore, it is worth emphasizing that the present approach is confined to periods of times which do not allow more than one or two oscillations of the field in the pulse envelope. It cannot be compared with previous approaches of multiphoton ionisation which requires many cycles [13, 14].

Now, if one replaces $\Psi(\mathbf{r},t)$ by $\chi_{\rm i}^{+}(\mathbf{r},t)$ in (1), one obtains the following equation

$$\left(H - \mathrm{i}\frac{\partial}{\partial t}\right)\chi_{\rm i}^{+} = \boldsymbol{\nabla}\phi_{\rm i}\cdot\boldsymbol{\nabla}L^{+}. \quad (10)$$

If the right-hand side is negligible, the CV state is close to an exact solution of (1). Therefore, a study of this term inform us about the accuracy of our approximation. Since $|\boldsymbol{\nabla}\phi_{\rm i}(\mathbf{r},t)|$ is roughly proportional to the orbital velocity, it decreases when the principal quantum number n increases. Then, we expect our spectra to be in better agreement with TDSE calculations for large values of n.

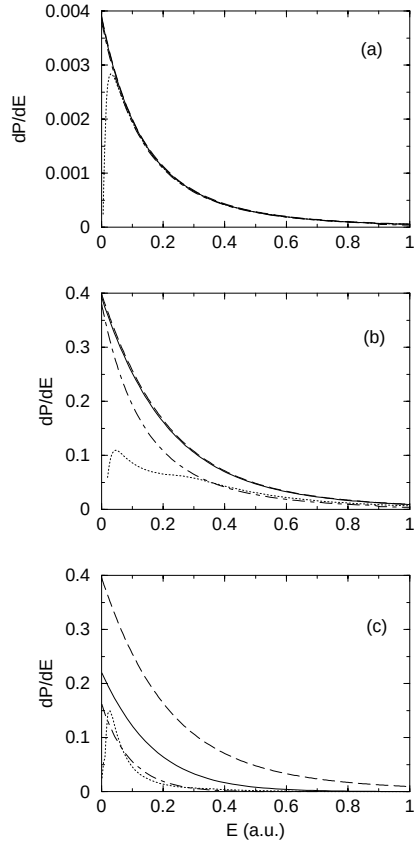

Fig. 1. Electron energy spectra obtained with Presnyakov's theory (dotted line), TDSE (full line), CV approach (long-dashed line) and FBA (dash-dotted line) for the atomic hydrogen in its ground state. The photon energy is 0.05 a.u. in all cases. (a) $E_0 = 0.1$, $\tau = 1$; (b) $E_0 = 1$, $\tau = 1$; (c) $E_0 = 0.1$, $\tau = 10$.

Finally, the integration of $|\langle\phi_{\rm f}^{-}|\chi_{\rm i}^{+}\rangle|^2$ over the direction of ejected electrons yields the electron energy distribution. Since the integral over r can be performed analytically, computer times never exceed a few seconds.

3 Results and discussion

The electron energy distribution given by the exact numerical solution TDSE reveals that the distribution predicted by Presnyakov's theory is incorrect close to the ionisation threshold. This behaviour is all the worse that the pulse duration is long or the intensity is high (see *e.g.* Fig. 1). Further, a singular point shows up when the time-dependent momentum of the continuum wave function is zero, *i.e.*, when $\mathbf{A}(t) = -\mathbf{k}$. The numerical integration close to this point is very lengthy. For long pulses, it is even longer than TDSE calculations. Furthermore, it is clear in Figure 1 that FBA predictions, which take no more than a few seconds, are always better than the predictions given by the Presnyakov's theory. We checked carefully that this situation is always the same with many different field parameters. Although the theory of Presnyakov seems to be well adapted to ionisation by swift ions [15], it appears not

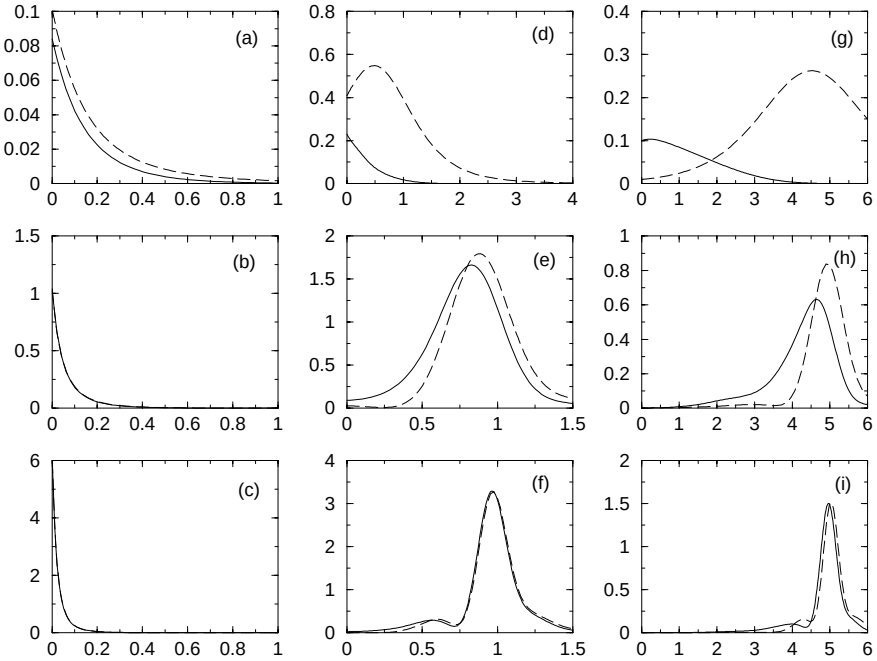

Fig. 2. Electron energy spectra predicted by TDSE (full line) and CV calculations (dashed line) for $E_0 = 0.1$ ($I_0 = 3.5 \times 10^{14}$ W/cm^2) and $\omega = 0.05$. 1st column: $\tau = 5$, 2nd column: $\tau = 30$, 3rd column: $\tau = 90$ ($\tau = 2.2$ fs). 1st row: H($1s$) targets, 2nd row: H($2s$) targets, 3rd row: H($3s$) targets.

to be adapted to the ionisation by ultra-intense and ultra-short laser pulses. It was the motivation to develop "the Coulomb-Volkov approach" which requires in our context much smaller computer times and which predicts the correct threshold behaviour (see below).

In this paper, calculations with the CV theory presented in Section 2.2 have been performed in some typical cases in order to find the domain where the method applies. Results for very short pulses ($\tau \leq 1$ a.u.) are not presented because TDSE and CV predictions are always identical in this case. Results given in Figures 2 and 3 aim at showing the smooth evolution of electron spectra when one increases the pulse duration up to values for which CV predictions depart from TDSE ones. Further, the maximum field amplitudes considered in Figures 2 and 3 are 0.1 and 1 a.u. (3.5×10^{16} W/cm^2) respectively.

Firstly, one notices that almost all spectra show a single well-defined peak. Since we are in cases where the external field has no time to oscillate, the electron is kicked out from the initial state by a field which exhibits essentially a classical aspect. Therefore, the energy given to the system is the "classical" energy:

$$E_{\mathrm{C}} = \frac{1}{2}\Big| - \int_0^\tau \mathrm{d}t E(t)\Big|^2 = \frac{\mathbf{A}(\tau)^2}{2}\cdot \tag{11}$$

More precisely, it can be shown analytically that the CV spectrum is peaked at the energy

$$E_{\mathrm{peak}} = E_{\mathrm{C}} + E_{\mathrm{i}} \tag{12}$$

where E_{i} is the energy of the initial state ($E_{\mathrm{i}} < 0$ with a threshold energy equal to zero). This result agrees with the conclusions of Bugacov *et al.* [16]. If $E_{\mathrm{C}} \geq -E_{\mathrm{i}}$, the peak shows up at the energy given by (12). Otherwise, the maximum ejection probability appears at the origin.

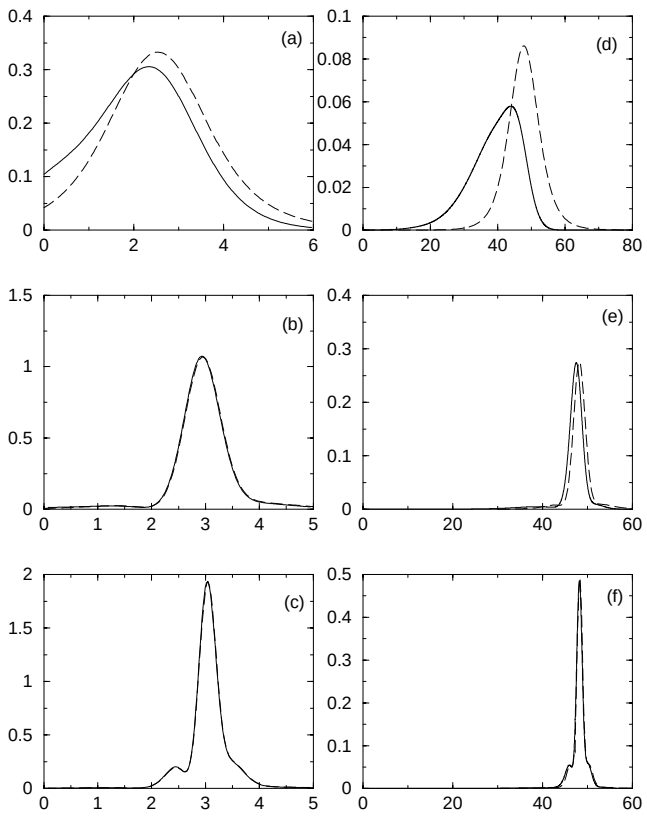

Fig. 3. Electron energy spectra predicted by TDSE (full line) and CV calculations (dashed line) for $E_0 = 1$ ($I_0 = 3.5 \times 10^{16}$ W/cm^2) and $\omega = 0.05$. 1st column: $\tau = 5$, 2nd column: $\tau = 20$. 1st row: H($1s$) targets, 2nd row: H($2s$) targets, 3rd row: H($3s$) targets.

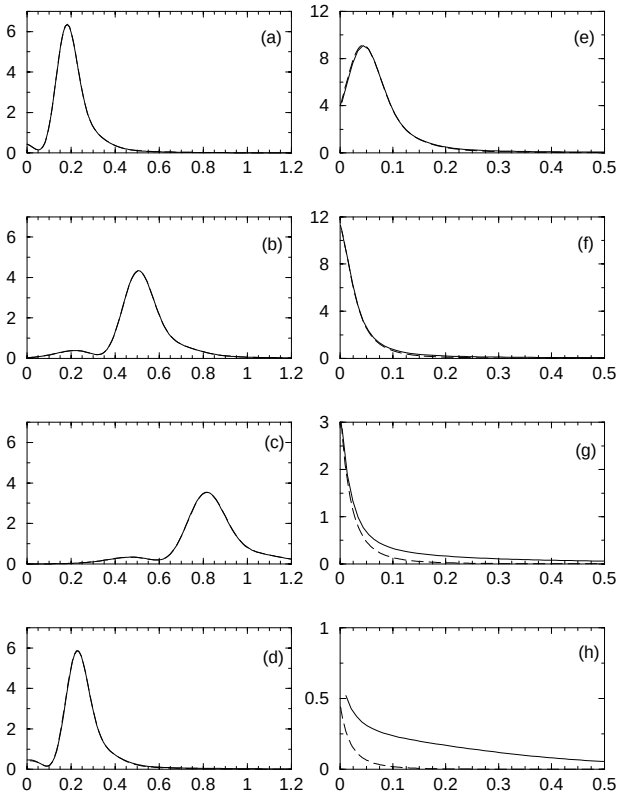

Fig. 4. A comparison between the electron energy spectra of TDSE (full line) and CV approach (dashed line) for H($3s$) with an increasing pulse duration to test the importance of the number of field oscillations. In all graphs $E_0 = 0.5$ and $\omega = 0.6$. Pulse lengths for (a) to (h) are 3, 5, 10, 15, 17, 18, 19, 20 respectively.

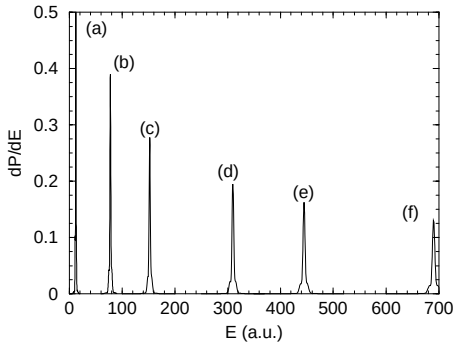

Fig. 5. CV predictions of the evolution of the energy spectrum when the pulse length τ is increased from 2 to 15. Laser field parameters are $E_0 = 5$ and $\omega = 0.05$ and targets are H($3s$) atoms. Spectra (a) to (f) are for values of $\tau = 2, 5, 7, 10, 12, 15$ respectively. One notices that the maximum of each peak, located at $E_C + E_i$ (see text), decreases smoothly with τ.

Now, it emerges from Figures 2 and 3 that CV results are all the worse that the interaction time τ is large. In addition, we can see that given τ, the CV predictions are better for large values of n. Thus we are led naturally to compare τ with the orbital period $T_n = 2\pi n^3$ of the initial bound state. One should have $\tau \leq T_n$ to consider that the sudden approximation applies. In fact, results given in Figure 2 show that accurate predictions with the CV approach are obtained for hydrogen targets when $\tau \leq T_n/2$.

For the laser intensities under consideration, the influence of the maximum field amplitude seems not to be important (the relative discrepancy between TDSE and CV spectra does not evolve significantly when the field is increased). For instance, the comparison between Figure 2b where the total ionisation probability is $P_i = 6.4 \times 10^{-2}$ (perturbative regime) and Figure 3b where $P_i = 0.993$ (saturation regime), clearly shows that the departure of CV from TDSE is the same. Therefore, one may assert that the CV method works in perturbative and non-perturbative regimes provided that the conditions of the sudden approximation are fulfilled.

Finally, results given in Figure 4 are a test of the CV approach when the field begins to oscillate. A large photon energy $\omega = 0.6$ is taken here to let the field oscillate during the pulse. The study is carried out with the atom in an initial $3s$ state because the orbital period is longer. Therefore, we can increase τ so that the field oscillates while keeping the system within the saturation regime. Indeed, Figures 2f and 3f show that for $\omega = 0.05$, the CV approach provides perfect predictions with an initial state $3s$ for $\tau \leq 20$ a.u. (*i.e*, $\tau \leq 0.5$ fs). Therefore, the discrepancy between TDSE and CV spectra, which appears in Figure 4, comes only from the fact that the field begins to oscillate. The difference becomes visible for $\tau \approx 18$ a.u. (Fig. 4f), *i.e.*, that is about 1.7 oscillation of the field. Thus, one may conclude that, with symmetric pulses, the CV approach applies when the field has less than 2 oscillations.

The power of the CV approach is particularly apparent with very strong fields. Predictions for $E_0 = 5$ a.u. ($\sim 10^{18}$ W/cm^2) and $\omega = 0.05$ are displayed in Figure 5 with a pulse duration τ increasing from 2 to 15 (we stop at this latest value of τ to prevent the ejected electron from reaching relativistic energies). Again, the initial state is the $3s$ state of hydrogen. We performed TDSE calculations on a IBM 590 workstation up to $\tau = 3$ with a computer time of about 10 hours (convergence parameters: the number of time steps is 131072, the box size is 200 a.u., the number and the order of B-splines are 800 and 7 respectively, the number of angular momentum is 190 in the length gauge) while CV calculations only take about 20 s. For $\tau = 3$, CV predictions are in a very good agreement with TDSE ones. TDSE calculations become much too long beyond $\tau = 3$ because the expansion of the wave function implemented in the code (based on a spherical symmetry) is not adapted anymore and the code barely converges.

Now, a smooth evolution of the CV electronic spectra appears in Figure 5 when τ increases up to 15. This situation supports the assumption that the CV approach is still reliable although there is now no reference theory to compare with it. Thus, a remarkable feature of our theory is to be able to provide some quantitative information where full numerical methods fail. Of course, atomic

hydrogen is the simplest atom to consider. However, for complex atoms, TDSE calculations with high fields which generate high energy electrons ($E \geq$ a few keV) become impossible. Therefore, it is of major interest to extend our approach to more complex atoms (alkaline and two-active electron atoms) to take advantage of its simplicity.

4 Conclusion

Our preliminary studies show that a very simple approach based on Coulomb-Volkov states may give reliable predictions on the ionisation of atoms by short and intense laser pulses. The method appears to be better adapted to cases where the principal quantum number of the initial state is large and the maximum of the field amplitude is high, a situation which excludes perturbative theories. Thus, our method is a theory which gives excellent predictions, even in non-perturbative conditions, as long as the interaction time is short enough to prevent oscillations of the field. Practically, field oscillation should not exceed 2 oscillations for a symmetric pulse.

It is worth noting that, in its domain of application, the Coulomb-Volkov approach is way faster than full numerical methods with the same accuracy. Moreover, in the case of strong fields where large energies are transferred to atoms, the present approach is a method that can predict accurately the energy distribution of ejected electrons at very low costs.

Finally, note that pulses even shorter than two optical cycles become now available [17], *i.e.,* with a total duration which matches the domain where the present approach applies.

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