

Spectral Analysis of the Photoionization of Atomic Hydrogen in Intense High-Frequency Laser Field: Numerical Simulation

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ABSTRACT: The present work aims at analyzing the dynamics of the photoionization process of a hydrogen atom -modelling a single active electron atom- interacting with intense high-frequency laser pulses. The choice of the numerical approach to be used for solving the time-dependent Schrödinger equation (TDSE) of an atom in a laser field is based on the fact that when the intensity of a femtosecond laser get higher, the electron after it reaches the continuum starts behaving as a free electron oscillating in the laser field, which leads to a strong oscillations in the electronic wave function, and then could affect the stability in numerical solutions of TDSE. Therefore a highly stable numerical methods are required for solving the TDSE of an atomic hydrogen in intense laser pulses, for this reason, we chose to use the three-point finite difference method for the spatial discretization of the wave function and the standard Peaceman-Rachford scheme coupled to an inverse iteration procedure for the function's propagation in time. Once the wave function obtained, a spectral analysis of the ejected electron based on the use of a window operator is performed to calculate the probability of ionization of a hydrogen atom by a high frequency laser field.

KEYWORDS: Spectral Analysis, Photoionization, Atomic Hydrogen, High-Frequency, Laser, Numerical Simulation.

1 INTRODUCTION

The study of the interaction of intense laser fields with atomic systems is subject of interest since the invention of laser since the early 60s. The development of high-power pulsed lasers during the past two decades has enabled us to reach of higher intensities to 10^{14} W/cm² and with pulse durations of a few femtoseconds (10^{-15} s). With such characteristics, the interaction of an intense laser fields with an atomic system leads to non-linear process where a large number of photons can be emitted or absorbed. Agostini et al. [4] discovered in 1979 that at sufficiently high intensities ($I > 10^{11}$ Wcm⁻²), the ejected electron can absorb photons in excess of the minimum number required for ionization to occur. This phenomenon is called "above-threshold ionization" (ATI). In the late 1980s the experimenters observed another phenomenon of noble gases interacting with intense, short-pulse laser fields: it is the high-order harmonic generation (HHG) [4,5].

So far, most theoretical efforts have been focused on solving the problem of a hydrogen atom, or more generally, a single-active-electron atomic system in a strong laser field for which exact calculations can be performed. As for multi-electron systems the theoretical calculations remains only an approximation.

The numerical treatment of the interaction of the hydrogenic atom with the intense, high frequency laser fields can not be treated within the framework of perturbation theory due to the interaction fields, which becomes comparable to the Coulomb field of hydrogen. Therefore, non-perturbative methods are required to explain most of the experimental observations. It is necessary in this case to solve the time-dependent Schrödinger equation (TDSE), which is usually found in non-perturbative theories. At the end of the interaction, our system is represented by the final wave function obtained from the numerical resolution of the TDSE, the analysis of the spectra of the ejected electrons whose ATI peaks were observed, can help us to calculate easily the probability of ionization of H(1s) by linearly polarized laser pulses.

2 CHARACTERISTICS OF THE LASER FIELD

The chosen laser field is characterized by its electric field $\vec{E}(\vec{r}, t)$ of peak amplitude $E_0 = 5 \cdot 10^{-3} \text{ a.u.}$ of frequency $\omega_0 = 0.57 \text{ u.a.}$ (corresponding to a period of $T_L = 2\pi/\omega_0 = 11.02 \text{ u.a.} \approx 0.25 \text{ fs}$) and modulated by an envelope $f(t)$. The total duration of the pulse t_{max} is defined by the following expression: $t_{\text{max}} = n_{\text{cycle}} T_L$, where n_{cycle} is the number of optical cycle of the laser field.

For a linearly polarized laser pulse, the electric field can be written in the dipole approximation as

$$\vec{E}(\vec{r}, t) = E_0 f(t) \sin(\omega_0 t) \vec{e}_z \quad (1)$$

where \vec{e}_z represents the polarization vector and $f(t)$ defines the temporal profile of the laser pulse (envelope), which is chosen to be trapezoidal. It rises linearly over five optical cycles, remains constant for thirty cycles then it is ramped linearly down over the final five cycles:

$$f(t) = \begin{cases} \frac{t}{5 \cdot T_L} & \text{si } 0 < t < 5 \cdot T_L \\ 1 & \text{si } 5 \cdot T_L < t \leq 35 \cdot T_L \\ -\frac{t}{5 \cdot T_L} + 8 & \text{si } 35 \cdot T_L < t < 40 \cdot T_L \end{cases}$$

The temporal evolution of the electric field defined with the expression (1) is then given in the following figure:

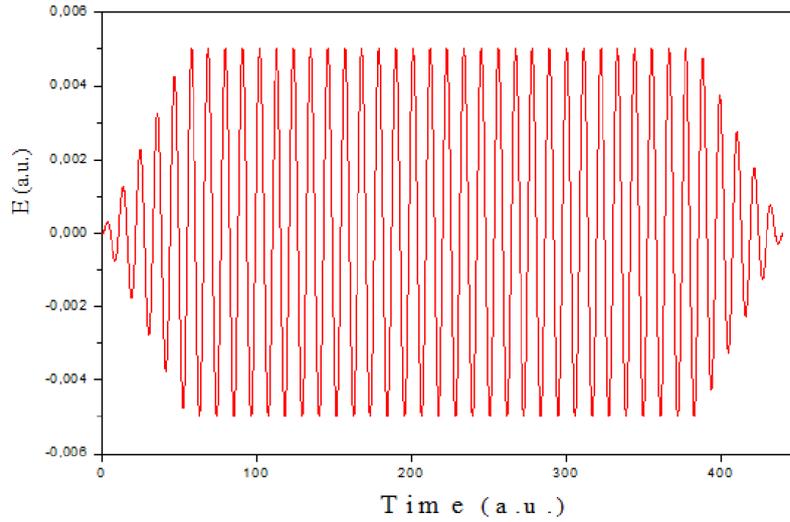


Figure 1: Temporal evolution of the electric field of the laser pulse with a frequency of $\omega_0 = 0.57 \text{ a.u.}$ (the photon energy of 15.5 eV) and a peak amplitude $E_0 = 5.10^{-3} \text{ a.u.}$ (Corresponding to a peak intensity of $8.77 \times 10^{11} \text{ W/cm}^2$) for 40 optical cycles (trapezoidal envelope)

3 NUMERICAL APPROACH

In addition to the analytical methods, different numerical methods have been developed to describe the interaction of the laser field with the active electron. Our numerical approach is to solve numerically the time-dependent Schrödinger equation (TDSE) in a non-perturbative way, namely

$$i \frac{\partial}{\partial t} \psi(\vec{r}, t) = (H_0 + H_{int}) \psi(\vec{r}, t), \quad (2)$$

where H_0 is the time-independent, non-relativistic field-free atomic Hamiltonian, which is written as follows:

$H_0 = E_c + E_p = -\frac{1}{2} \Delta - \frac{1}{r}$, with E_c the kinetic energy of the electron, and E_p the potential energy. The term H_{int} represents the time-dependent Hamiltonian describing the interaction of the atom with the laser field given by $\vec{E}(t) \cdot \vec{r}$ in length gauge or $\vec{A}(t) \cdot \vec{p}$ in velocity gauge with $\vec{E}(t)$ defined in (1) is the electric field of the laser and $\vec{A}(t)$ is the potential vector.

If we limit ourselves to laser pulse linearly polarized along the z-axis, the magnetic quantum number m of the electron is conserved, the time-dependent Schrödinger equation describing the atom in a laser field in the length gauge is given by (in atomic units)

$$i \frac{\partial}{\partial t} \psi(\vec{r}, t) = \left\{ -\frac{1}{2} \left(\frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial}{\partial r} \right) + \frac{1}{r^2} \bar{L}^2 \right) - \frac{1}{r} - E_0 z f(t) \sin(\omega_0 t) \right\} \psi(\vec{r}, t), \quad (3)$$

where the position \vec{r} is expressed using the spherical coordinates (r, θ, φ) .

We can separate the radial part of the wave function from its angular part by expanding it in spherical harmonics

$$\psi(r_i, \theta, \varphi, t) = \sum_{\ell=0}^L \Phi_{\ell}(r_i, t) Y_{\ell}^0(\theta, \varphi), \quad \Phi_{\ell}(r_i, t) = \frac{R_{\ell}^i(t)}{r_i} \quad (4)$$

The finite difference representation at three points can be employed for a discretized representation of the kinetic energy, the field-free Hamiltonian H_0 becomes

$$\begin{aligned}
 H_0 \mathcal{W}(r_i, \theta, \varphi, t) &= \frac{1}{r_i} \sum_{\ell} Y_{\ell}^0(\theta, \varphi) \left\{ -\frac{1}{2(\Delta r)^2} (c_i R_{\ell}^{i+1}(t) + c_{i-1} R_{\ell}^{i-1}(t) - 2d_i R_{\ell}^i(t)) + \left(\frac{\ell(\ell+1)}{2r_i^2} - \frac{1}{r_i} \right) R_{\ell}^i(t) \right\} \\
 &= \frac{1}{r_i} \sum_{\ell} Y_{\ell}^0(\theta, \varphi) (H_0 R(t))_{\ell}^i
 \end{aligned} \tag{5}$$

with $r_i = (i - 0.5)\Delta x$ and the coefficients c_i and d_i are defined by [6]

$$c_i = \frac{i^2}{i^2 - 1/4}, \quad d_i = \frac{i^2 - i + 1/2}{i^2 - i + 1/4}.$$

We see that the Hamiltonian matrix H_0 is diagonal in the angular momentum quantum number ℓ and is tridiagonal in the grid index i , therefore the non-zero elements are given by

$$H_{0,\ell,\ell'}^{i,i} = \delta_{\ell,\ell'} \begin{cases} -\frac{c_i}{2(\Delta r)^2} & i = i+1 \\ -\frac{c_{i-1}}{2(\Delta r)^2} & i = i-1 \\ \frac{d_i}{(\Delta r)^2} + \frac{\ell(\ell+1)}{2r_i^2} - \frac{1}{r_i} & i = i \end{cases} \tag{6}$$

Substituting Equations (4) and (5) into the TDSE (3), we find that

$$i \frac{\partial R_{\ell}^i(t)}{\partial t} = (H_0 R(t))_{\ell}^i + \sum_{\ell'} H_{\ell\ell'} R_{\ell'}^i(t), \tag{7}$$

with $H_{\ell\ell'}$ are the matrix elements of the interaction Hamiltonian

$$H_{\ell\ell'} = \langle Y_{\ell}^0(\theta, \varphi) | H_{int} | Y_{\ell'}^0(\theta, \varphi) \rangle \tag{8}$$

The interaction Hamiltonian in the length gauge is given by

$$H_{int} = -zE(t) = -E(t)r \cos \theta, \tag{9}$$

θ is the angle between the vector r and the z-axis.

The matrix elements of the interaction Hamiltonian in the angular momentum basis is given by

$$\begin{aligned}
 \langle \ell 0 | H_{int} | \ell' 0 \rangle &= -E(t)r_i \langle \ell 0 | \cos \theta | \ell' 0 \rangle \\
 &= -E(t)r_i \left(\frac{\ell+1}{\sqrt{(2\ell+1)(2\ell+3)}} \delta_{\ell,\ell+1} + \frac{\ell}{\sqrt{(2\ell+1)(2\ell-1)}} \delta_{\ell,\ell-1} \right), \\
 &= -\delta_{\ell,\ell'} E(t)r_i \begin{cases} a_{\ell}, & \ell' = \ell+1 \\ a_{\ell-1}, & \ell' = \ell-1 \end{cases}
 \end{aligned} \tag{10}$$

where the coefficients a_{ℓ} are given by

$$a_{\ell} = \frac{\ell+1}{\sqrt{(2\ell+1)(2\ell+3)}} \tag{11}$$

This matrix H_{int} is then tridiagonal in the angular momentum quantum number ℓ and diagonal in the grid index i .

Substituting Equation (9) into Equation (6), we can then rewrite the TDSE in the form

$$i \frac{\partial R_\ell^i(t)}{\partial t} = -\frac{1}{2(\Delta r)^2} (c_i R_\ell^{i+1}(t) + c_{i-1} R_\ell^{i-1}(t) - 2d_i R_\ell^i(t)) + \left(\frac{\ell(\ell+1)}{2r_i^2} - \frac{1}{r_i} \right) R_\ell^i(t) - E_0 r_i f(t) \sin(\omega_0 t) \{ a_\ell \delta_{\ell',\ell+1} + a_{\ell-1} \delta_{\ell',\ell-1} \} R_\ell^i(t) \quad (12)$$

Since both the matrix H_0 and the matrix $H_{int}(t)$ are tridiagonal, we can use the implicit method of Peaceman-Rachford, which consists in replacing the differential equation by two discretized equations (tridiagonal) used for periods $\frac{\Delta t}{2}$, namely

$$R_\ell^i(t + \Delta t) = \left[I + iH_0 \frac{\Delta t}{2} \right]^{-1} \left[I + iH_I \frac{\Delta t}{2} \right]^{-1} \left[I - iH_0 \frac{\Delta t}{2} \right] \left[I - iH_I \frac{\Delta t}{2} \right] R_\ell^i(t) \quad (13)$$

Let $\left[I - iH_0 \frac{\Delta t}{2} \right] \left[I - iH_I \frac{\Delta t}{2} \right] R_\ell^i(t) = \chi_\ell^i(t) \quad (14)$

$$\underbrace{\left[I + iH_0 \frac{\Delta t}{2} \right]}_{tridiagonale} \xi_\ell^i(t + \Delta t) = \chi_\ell^i(t) \quad (15)$$

Using the Routine Tridag for solving a tridiagonal system, we can calculate $\xi_\ell^i(t + \Delta t)$.

Finally we must solve the following system to find $R_\ell^i(t + \Delta t)$

$$\underbrace{\left[I + iH_I \frac{\Delta t}{2} \right]}_{tridiagonal} R_\ell^i(t + \Delta t) = \xi_\ell^i(t + \Delta t) \quad (16)$$

In our simulations, we took $\Delta r = 0.1 a.u.$, $\Delta \tau = 0.1 a.u.$ and the ground state of the hydrogen atom $R_\ell^i(t = 0) = 2r_i e^{-r_i}$ as the initial state of the system.

4 SPECTRUM OF THE EJECTED ELECTRON

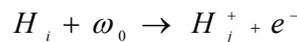
4.1 WINDOWS OPERATOR TECHNIQUE

For laser intensities of about 10^{12} W/cm^2 , the ejected electron can absorb a number of photons in excess of the minimum number required for ionization; by having a number N of photons absorbed, then additional photons S above the threshold, the bounded electron thus acquired a kinetic energy equal to

$$E_c = (N + S) \hbar \omega_0 - |E_i| \quad (17)$$

where E_i is the ionization energy, that is the minimum energy required for the electron initially located in the ground state ($n = 1$) to be no longer bound to the atom.

In our case, a linearly polarized photon, having an energy of $\omega_0 = 0.57 a.u. > E_i = \frac{e^2}{8\pi a_0 \epsilon_0} \approx 0.5 a.u.$, remove the electron from the hydrogen atom, therefore the dominant ionization process will be photoionization ($N = 1$), it can be summarized by the following reaction



At the end of the pulse laser, i.e. at time t_{\max} , our atomic system was represented by final-state wave function $\psi_f(r, t_{\max})$ from which we can deduce the energy spectrum of the ejected electrons by using an adequate method, which does not require the computation of all the eigenstates of the system. It is based on the use of a window operator $\hat{F}(E_k, n, \gamma)$ [7]:

$$\hat{F}(E_k, n, \gamma) = \frac{\gamma^{2^n}}{(E_k)^{2^n} + \gamma^{2^n}}, \tag{18}$$

with n is the order of the window operator, and γ is the parameter, which determines the spectral width of the window, i.e. the energy resolution of the analysis. A good value of γ should not be too big if you want to be able to extract information such as ATI spectrum peaks.

Using the window operator (see Eq. 18) we can determine the energy spectrum of photoelectrons ejected from hydrogen, such as the total probability of finding the electron energy (in the final state) within the interval $E_k \pm \gamma$

$$P(E_k, n, \gamma) = \langle \psi_f | \frac{\gamma^{2^n}}{(H_0 - E_k)^{2^n} + \gamma^{2^n}} | \psi_f \rangle, \tag{19}$$

$$= \langle \chi | \chi \rangle$$

with

$$|\chi\rangle = \frac{\gamma^{2^{n-1}}}{(H_0 - E_k)^{2^{n-1}} + i\gamma^{2^{n-1}}} |\psi_f\rangle \tag{20}$$

To satisfy a good energy resolution, we choose $n = 2$ [6], thus equation (20) becomes

$$[(H_0 - E_k)^2 + i\gamma^2] |\chi\rangle = \gamma^2 |\psi_f\rangle \tag{21}$$

$$\underbrace{[(H_0 - E_k) + \sqrt{i}\gamma]}_{\text{tridiagonal}} \underbrace{[(H_0 - E_k) - \sqrt{i}\gamma]}_{\text{tridiagonal}} |\chi\rangle = \gamma^2 |\psi_f\rangle \tag{22}$$

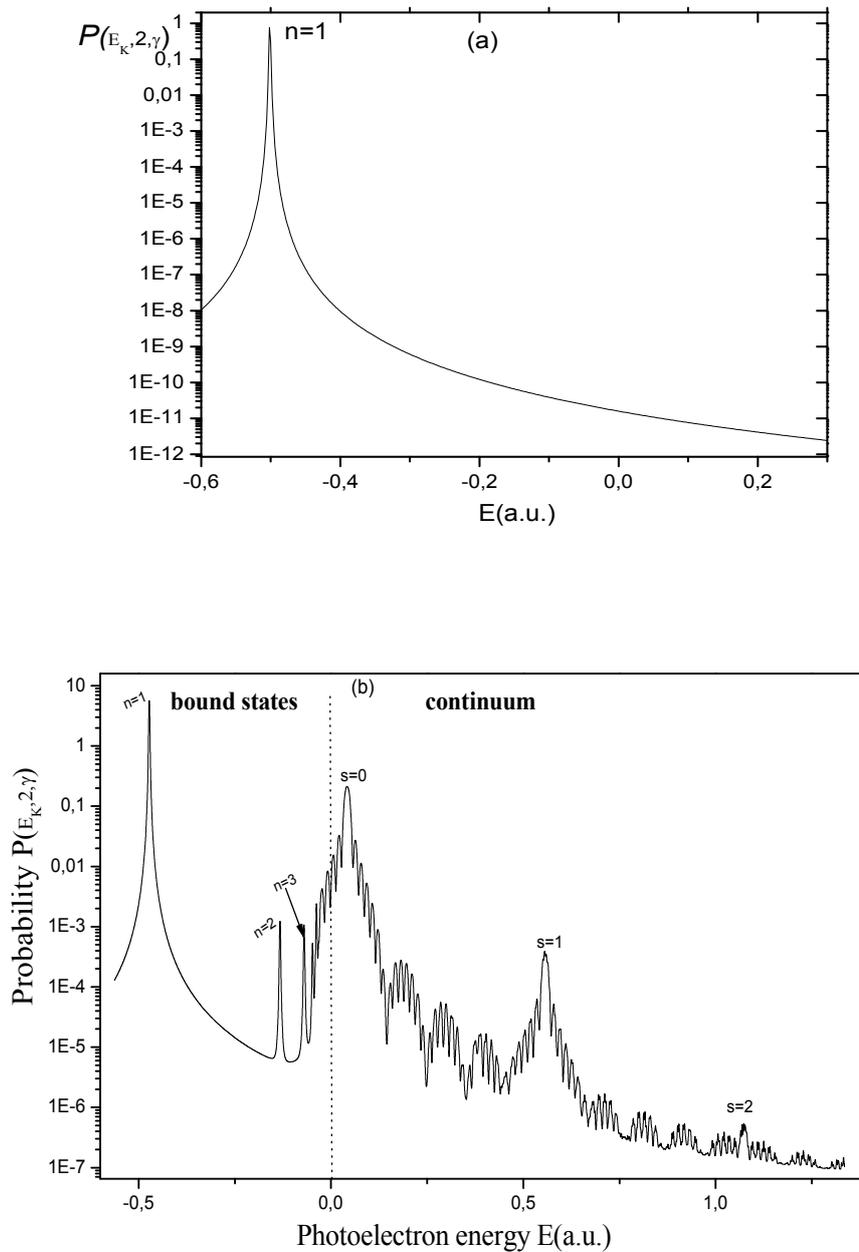


Figure 2: Probability $P(E_k, n, \gamma)$ for $n = 2$, $\gamma = 10^{-3}$ a.u., in Logarithmic scale as function of the photoelectron energy. We use $L_{\max} = 8$ and a forty-cycle laser pulse of photon energy $\omega_0 = 0.57$ a.u. and maximum amplitude : (a) $E_0 = 0$ a.u. (b) $E_0 = 5.10^{-3}$ a.u..

Figure (2) shows the probability $P(E_k, n, \gamma)$ on a logarithmic scale. Peaks whose energy $E < 0$ correspond to the bound states of the atom, while in the continuum, there is well positioned three peaks at 0.0699 a.u., 0.639 and 1.209 a.u. respectively, which are separated by one photon energy $\omega_0 = 0.57$ a.u. This agrees fairly well with the expected theoretical value. Indeed, the first peak of the spectrum corresponds to the ionization of hydrogen from its 1s ground state by absorbing

a photon of energy $\omega_0 = 0.57 \text{ a.u.}$, while the second and third peaks are related to the ATI. The widths and shapes of these peaks are consistent with the Fourier transform of the laser pulse shape, as is expected if no resonance enhancement is involved.

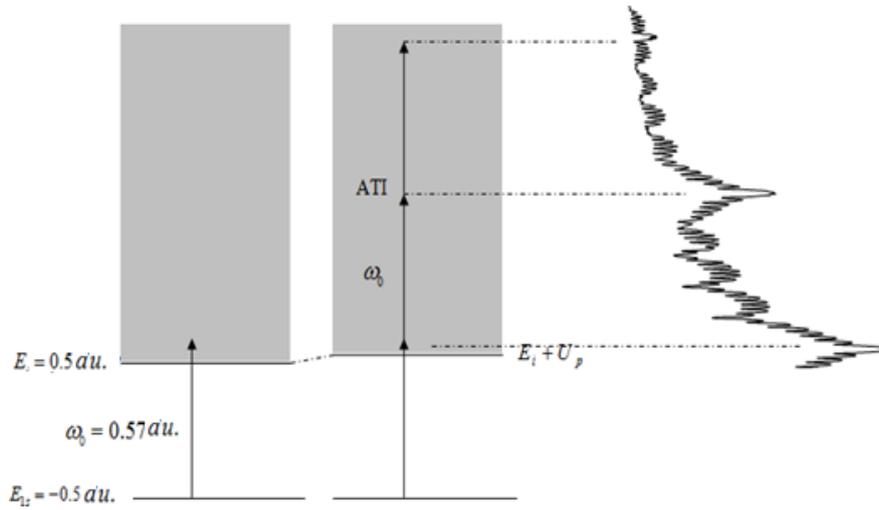


Figure 3: Shifting of ATI peaks due to ponderomotive effects, which cause an increase in the ionization potential value.

The ATI peak position is seen to shift with a value of U_p toward lower energies which results in a change in the ionization potential of the atom. U_p can be interpreted as the cycle-averaged kinetic energy of a quivering electron in a laser and can be written in a.u. as:

$$U_p = \frac{I(W/cm^2)}{4\omega^2(a.u.)I_0(W/cm^2)}, \quad (23)$$

with $I_0(W/cm^2) = 3.5 \times 10^{16} W/cm^2$.

The kinetic energy of the ionized electron (17) can be rewritten as follows

$$E_c = (N+S)\hbar\omega_0 - (|E_i| + U_p) \quad (24)$$

This last expression shows that the released electron energy depends not only on the number of the absorbed photons, but also on the value of the field at its release. Therefore, the ATI peaks have a certain width $\Delta E \simeq 2 \times 10^{-2} \text{ a.u.}$ (the photon energy divided over the length of the pulse), this value is in good agreement with those extracted from Figure (2).

5 PROBABILITY OF TOTAL IONIZATION

We can calculate the total ionization probability from the ATI spectrum:

$$\begin{aligned} p_{ion} &= \int_0^{E_{max}} \frac{dP(E)}{dE} dE = \int_0^{E_{max}} \rho(E) dE \\ &\approx \sum_k \rho(E_k) \Delta E_k \end{aligned}, \quad (25)$$

where $\rho(E_k)$ is the probability density of finding the electron ejected with an energy close to E_k .

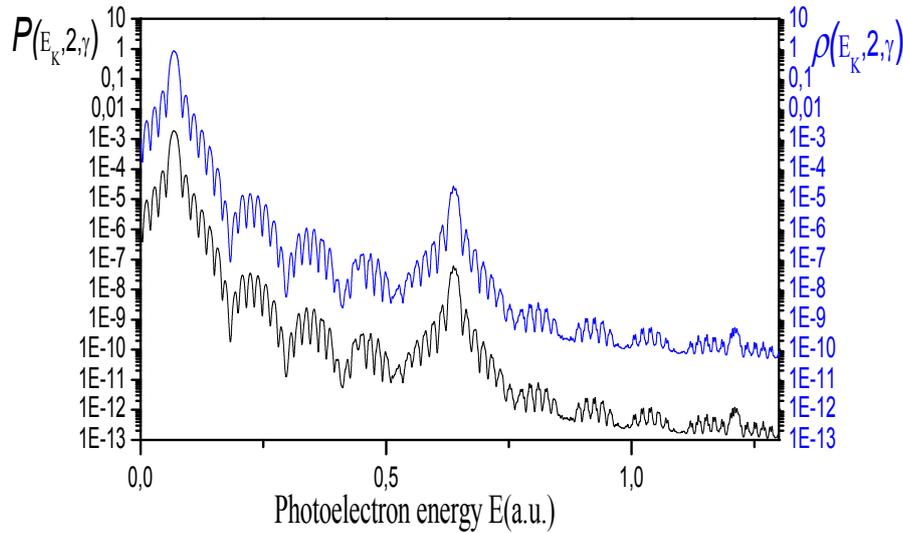


Figure 4: probability density of the free electrons $\rho(E_k, 2, \gamma)$ (blue) and the probability $P(E_k, n, \gamma)$ (black), with a spectral width $\gamma = 10^{-3}$ and with the same laser pulse parameters $(\omega_0, E_0, n_{c.o.})$ as in Fig. (2).

In our system there is only one electron that is involved in the process, the final state can then be developed in the basis of eigenvectors of H_0 :

$$|\psi_f\rangle = \sum_{|\ell\rangle} c_\ell |\ell\rangle + \int_{e>0} de c(e) |e\rangle \tag{26}$$

Thus we can write $P(E_k, n, \gamma)$ (Equation 19) as a sum over the bound states and an integrand over the continuum:

$$P(E_k, n, \gamma) = P_\ell + P_c$$

$$= \sum_{|\ell\rangle} |c_\ell|^2 \hat{F}(E_k - E_\ell, n, \gamma) + \int_{e>0} de |c(e)|^2 \hat{F}(E_k - e, n, \gamma) \tag{27}$$

It corresponds to the total probability of locating the electron either in the bound states of the atom or in the states of the continuum.

In the region $E > 0$, equation (27) becomes:

$$P(E_k, n, \gamma) \approx \int_{e>0} de |c(e)|^2 \hat{F}(E_k - e, n, \gamma) \tag{28}$$

If the value of γ is chosen small enough such that $c(e)$ does not change too much on a large area of energy [5], (28) can be written [8]:

$$\begin{aligned}
 P(E_k, n, \gamma) &\approx |c(E)|^2 \int_{e>0} de \hat{F}(E_k - e, n, \gamma) \\
 &= |c(E)|^2 \frac{\pi}{n} \operatorname{csc}\left(\frac{\pi}{2n}\right) \gamma
 \end{aligned} \tag{29}$$

This formula makes the link between the probability density $|c(E)|^2$ and the probability $P(E_k, n, \gamma)$:

$$\rho(E_k, n, \gamma) = \frac{P(E_k, n, \gamma)}{\frac{\pi}{n} \operatorname{csc}\left(\frac{\pi}{2n}\right) \gamma} \tag{31}$$

We can then convert the ATI spectrum in a probability density, by using equation (31) the conversion factor is then

$\frac{\pi}{2} \operatorname{csc}\left(\frac{\pi}{4}\right) \gamma$ (for $n = 2$). The ionization probability (Equation 27) p_{ion} can be approximated by the following formula:

$$p_{ion} \approx \frac{1}{\frac{\pi}{n} \operatorname{csc}\left(\frac{\pi}{2n}\right) \gamma} \sum_k P(E_k, n, \gamma) \Delta E_k \tag{32}$$

As part of our calculation, the expression (32) gives us a probability of ionization 0.013983 au, this value is close to that calculated from the projection of the final wave function on the bra of the initial (fundamental) state $\langle \varphi_{1s} |$ such as:

$$p_{ion}(t) \approx 1 - \left| \langle \varphi_{1s} | \psi(t) \rangle \right|^2 \tag{33}$$

we notice from Figure (5), that at the end of the interaction $p_{ion} \approx 1 - P_{n=1} = 0.01397 \text{ a.u.}$

A second procedure for calculating the total ionization probability is given by calculating the total probability for an electron to remain in the bounded states after the interactions:

$$P_{\text{états liés}} = \sum_n \left| \langle \varphi_n | \psi(t_{\max}) \rangle \right|^2 \tag{34}$$

the total probability of ionization of the system is:

$$p_{ion} = 1 - P_{\text{états liés}} \tag{35}$$

Using Figure (2), we can easily extract the amount of probability for each bound state $P_{\text{états liés}}$ such as: the result for $n = 1$ is in good agreement with that of the ground state population at the end of the interaction $\left| \langle \varphi_n | \psi(t_{\max}) \rangle \right|^2$, while the contributions of $n \geq 2$ are found to be of 10^{-7} u.a. of magnitude, which allows us to neglect them in our study.

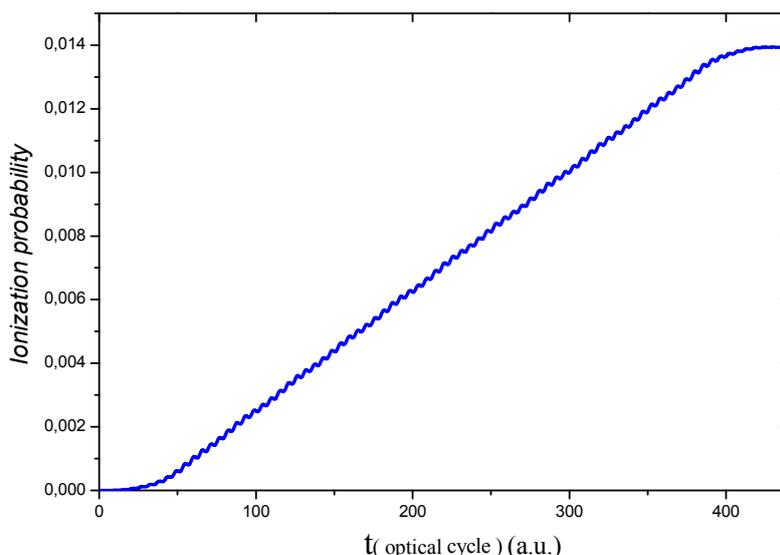


Figure 5: Ionization probability, $1 - \left| \langle \varphi_{1s} | \psi(t) \rangle \right|^2$, versus time for the same laser field parameters as above.

6 CONCLUSION

We have presented numerical methods that we have used to solve the time dependent Schrödinger equation for a hydrogen atom in presence of a strong and short laser field. The obtained wave function was analyzed in energy by means of a window operator which allowed us to analyze the energy spectra of the ejected electron. We observed typical ATI peaks separated by one photon energy and we evaluated the probability of the photoionization of the hydrogen in such extreme conditions.

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