Dynamic interference of photoelectrons in multiphoton ionization of atoms by short laser pulses

N. S. Simonović, D. B. Popović and A. Bunjac

Institute of Physics, University of Belgrade, P.O. Box 57, 11001 Belgrade, Serbia

1. The three-level model

The sequential two-photon ionization of the hydrogen atom by a short laser pulse is studied using the three-level model. The atom, which is initially in its *ground state* $|I\rangle = |1\mathrm{s}\rangle$ (we sete $E_I = 0$), is resonantly excited into an *intermediate state* $|R\rangle = |n\mathrm{p}\rangle$ $(n \geq 2)$ by absorption of a single photon of energy $\omega = E_R = I_p - \frac{1}{2n^2}$ and subsequently ionized by a second photon into a final electron *continuum state* $|F\rangle$ of energy $E_F = I_p + \varepsilon$. Here, $I_p = \frac{1}{2}$ is the ionization potential of the atom and $\varepsilon = k^2/2$ is the kinetic energy of the emitted photoelectron (atomic units are used throughout).

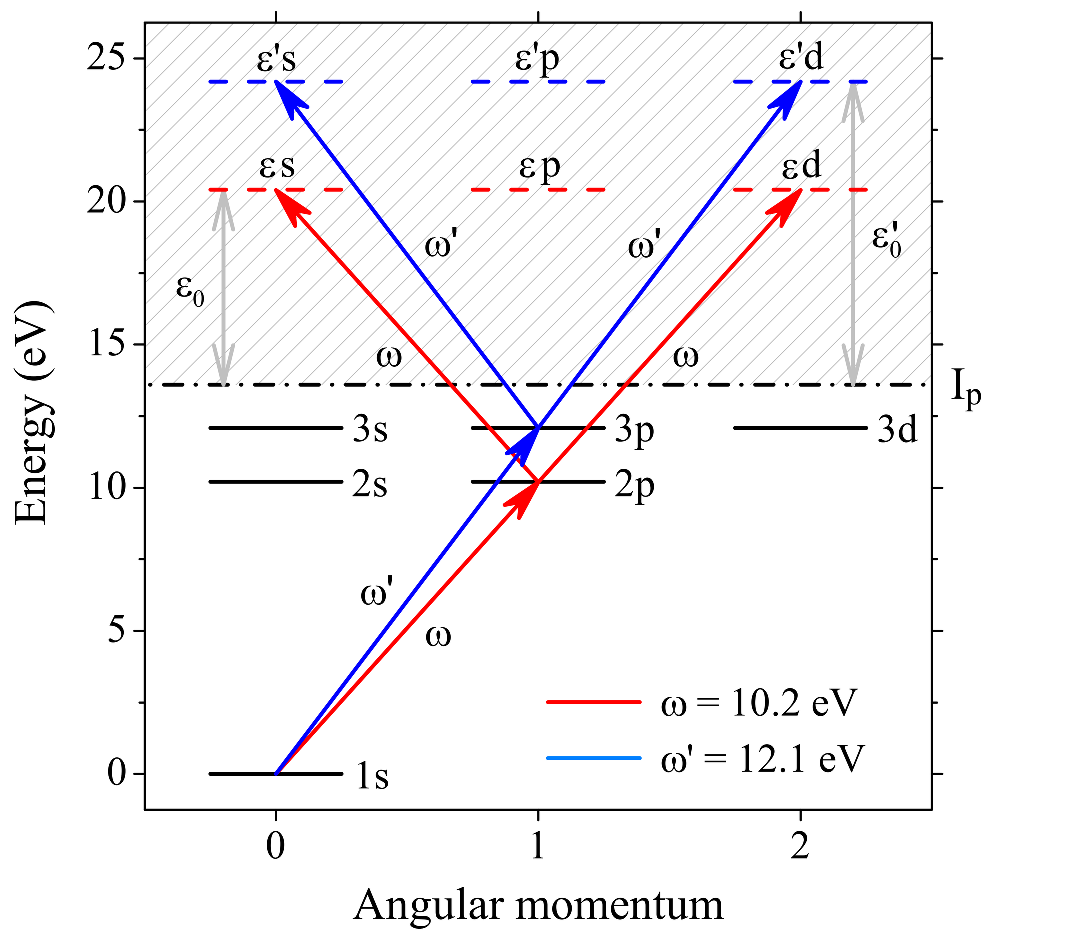

Figure 1. Energy level scheme of the hydrogen atom and the two-photon absorption paths for transitions from the ground (1s) state to the final continuum states (εs and εd) via one-photon resonant excitation of a np state ($n = 2, 3$).

The final state wave function at photoionization of the hydrogen-like atom satisfies the incoming wave boundary condition and can be written as the partial wave expansion

$$\psi^{(-)}_{\mathbf{k}}(\mathbf{r}) = \sqrt{\frac{2}{\pi}}\frac{1}{k}\sum_{l,m} i^l e^{-i\sigma_l}\frac{u_l(k,r)}{r}Y_{lm}(\Omega)Y^*_{lm}(\Omega_{\mathbf{k}}), \quad (1)$$

where $\sigma_l = \arg\Gamma(l + 1 + i\eta)$ is the Coulomb phase shift with the Sommerfeld parameter $\eta = -Z/k$ (for hydrogen $Z = 1$) and the radial function $u_l(k, r)$ is given by the regular Coulomb function $F_l(\eta, kr)$ [1].

Applying *a linearly polarized laser pulse*

$$\mathcal{E}(t) = \mathcal{E}_0\, g(t)\cos\omega t \quad (2)$$

with envelope $g(t)$, *the state of the atom at time t* reads

$$|\psi(t)\rangle = a_I(t)|I\rangle + a_R(t)e^{-i\omega t}|R\rangle + \int a_{\mathbf{k}}(t)e^{-2i\omega t}|\mathbf{k}\rangle \mathrm{d}^3\mathbf{k}, \quad (3)$$

where $a_I(t)$, $a_R(t)$ and $a_{\mathbf{k}}(t)$ are the *time-dependent amplitudes* for the population of states $|I\rangle$, $|R\rangle$ and $|F\rangle = |\mathbf{k}\rangle$ (represented by Eq. (1)), respectively. The stationary states $|R\rangle$ and $|F\rangle$ have been dressed by multiplying with the phase factors $e^{-i\omega t}$ and $e^{-2i\omega t}$ to simplify the equations of motion.

Inserting Eq. (3) into the time-dependent Schrödinger equation for the total Hamiltonian $\hat{H}(t) = \hat{H}_0 + \hat{W}(t)$, where $\hat{H}_0$ is the Hamiltonian of the field-free atom and the term $\hat{W}(t) = \hat{z}\mathcal{E}(t)$ describes *the atom-field interaction*, and applying the rotating wave approximation, we obtain the following set of *equations for the amplitudes*

$$\begin{aligned} i\dot{a}_I &= \frac{1}{2}D^*\mathcal{E}_0\, g(t)\, a_R(t),\\ i\dot{a}_R &= \frac{1}{2}D\,\mathcal{E}_0\, g(t)\, a_I(t) + \frac{1}{2}\mathcal{E}_0\, g(t)\int d^*(\mathbf{k})\, a_{\mathbf{k}}(t)\,\mathrm{d}^3\mathbf{k},\\ i\dot{a}_{\mathbf{k}} &= \frac{1}{2}d(\mathbf{k})\,\mathcal{E}_0\, g(t)\, a_R(t) + \left(I_p + \frac{k^2}{2} - 2\omega\right)a_{\mathbf{k}}(t). \end{aligned} \quad (4)$$

Here $D = \langle R|\hat{z}|I\rangle$ and $d(\mathbf{k}) = \langle F|\hat{z}|R\rangle$ are the dipole transition matrix elements for the excitation of the intermediate state and for its subsequent ionization, respectively.

2. Local approximation

Using a formal solution of the third of Eqs. (4)

$$a_{\mathbf{k}}(t) = -\frac{i}{2}\mathcal{E}_0\, d(\mathbf{k})\int_{-\infty}^{t} e^{-i(I_p + k^2/2 - 2\omega)(t-t')} g(t')\, a_R(t')\,\mathrm{d}t' \quad (5)$$

and the *local approximation* [2] which assumes that the main contribution of the integral over t' stems from the times around $t' \sim t$, the last term in the second of Eqs. (4) reduces to

$$\frac{1}{2}\mathcal{E}_0\, g(t)\int d^*(\mathbf{k})\, a_{\mathbf{k}}(t)\,\mathrm{d}^3\mathbf{k} = -\pi i\frac{|d_{\varepsilon_0}|^2\mathcal{E}_0^2}{4}g^2(t)\, a_R(t), \quad (6)$$

where $|d_\varepsilon|^2 = k\int_{\Omega_{\mathbf{k}}}|d(\mathbf{k})|^2\mathrm{d}\Omega_{\mathbf{k}}$, $k = \sqrt{2\varepsilon}$ and $\varepsilon_0 = 2\omega - I_p$ (see Fig. 1). Finally, one obtains the set of equations [3]

$$\begin{aligned} i\dot{a}_I &= \frac{1}{2}D^*\mathcal{E}_0\, g(t)\, a_R(t),\\ i\dot{a}_R &= \frac{1}{2}D\mathcal{E}_0\, g(t)\, a_I(t) - \frac{i}{2}\Gamma g^2(t)\, a_R(t),\\ i\dot{a}_\varepsilon &= \frac{1}{2}d_\varepsilon\,\mathcal{E}_0\, g(t)\, a_R(t) + (I_p + \varepsilon - 2\omega)a_\varepsilon(t), \end{aligned} \quad (7)$$

where $a_\varepsilon(t) = d_\varepsilon a_{\mathbf{k}}(t)/d(\mathbf{k})$ and

$$\Gamma = 2\pi\left|\frac{d_{\varepsilon_0}\mathcal{E}_0}{2}\right|^2 \quad (8)$$

is *the ionization rate* of the intermediate resonant state $|R\rangle$. The imaginary term $-\frac{i}{2}\Gamma g^2(t)$ describes the losses of the population of the intermediate state by the ionization into all final electron continuum states $|F\rangle$.

3. Results

The system of Eqs. (7) was solved numerically for two values of the photon energy ω which fit the excitation energies of: (i) 2p state ($\omega = E_R = 3/8\,\mathrm{a.u.} = 10.2\,\mathrm{eV}$) and (ii) 3p state ($\omega = E_R = 4/9\,\mathrm{a.u.} = 12.1\,\mathrm{eV}$), choosing a gaussian laser pulse

$$g(t) = e^{-t^2/\tau^2} \quad (9)$$

of $\tau = 30\,\mathrm{fs}$ duration and different intensities $I_0 = \mathcal{E}_0^2/(8\pi\alpha)$ (in the TW/cm^2 domain, $\alpha = 1/137$). The computed dipole transition matrix elements for the excitation and ionization are: (i) $D = 0.744936\,\mathrm{a.u.}$, $d_{\varepsilon_0} = 0.407759\,\mathrm{a.u.}$ and (ii) $D = 0.298311\,\mathrm{a.u.}$, $d_{\varepsilon_0} = 0.153418\,\mathrm{a.u.}$

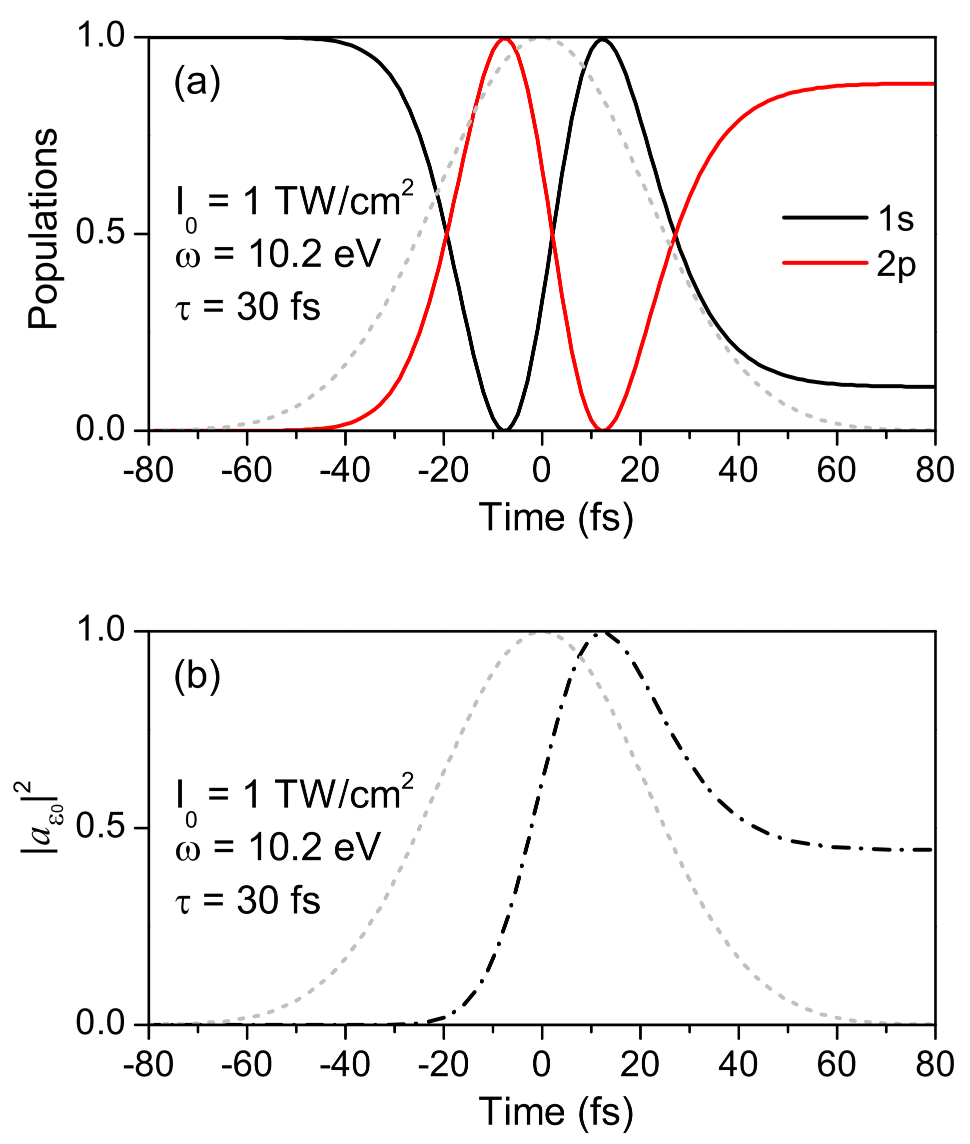

Figure 2. (a) The populations of the ground state (1s) and the intermediate resonant 2p state, calculated as $|a_I(t)|^2$ and $|a_R(t)|^2$, respectively, at the sequential two-photon ionization of hydrogen by a gaussian laser pulse of 1 TW/cm^2 peak intensity, 30 fs duration and the carrier frequency of $\omega = 3/8\,\mathrm{a.u.} = 10.2\,\mathrm{eV}$ which fits to the energy of 1s → 2p transition. (b) The photoelectron yield at $\varepsilon = \varepsilon_0$ in the same ionization process, calculated as $|a_{\varepsilon_0}(t)|^2$. The dashed gray line represents the envelope of the laser pulse. For the chosen peak intensity the populations perform approx. 1.5 Rabi cycles during the pulse.

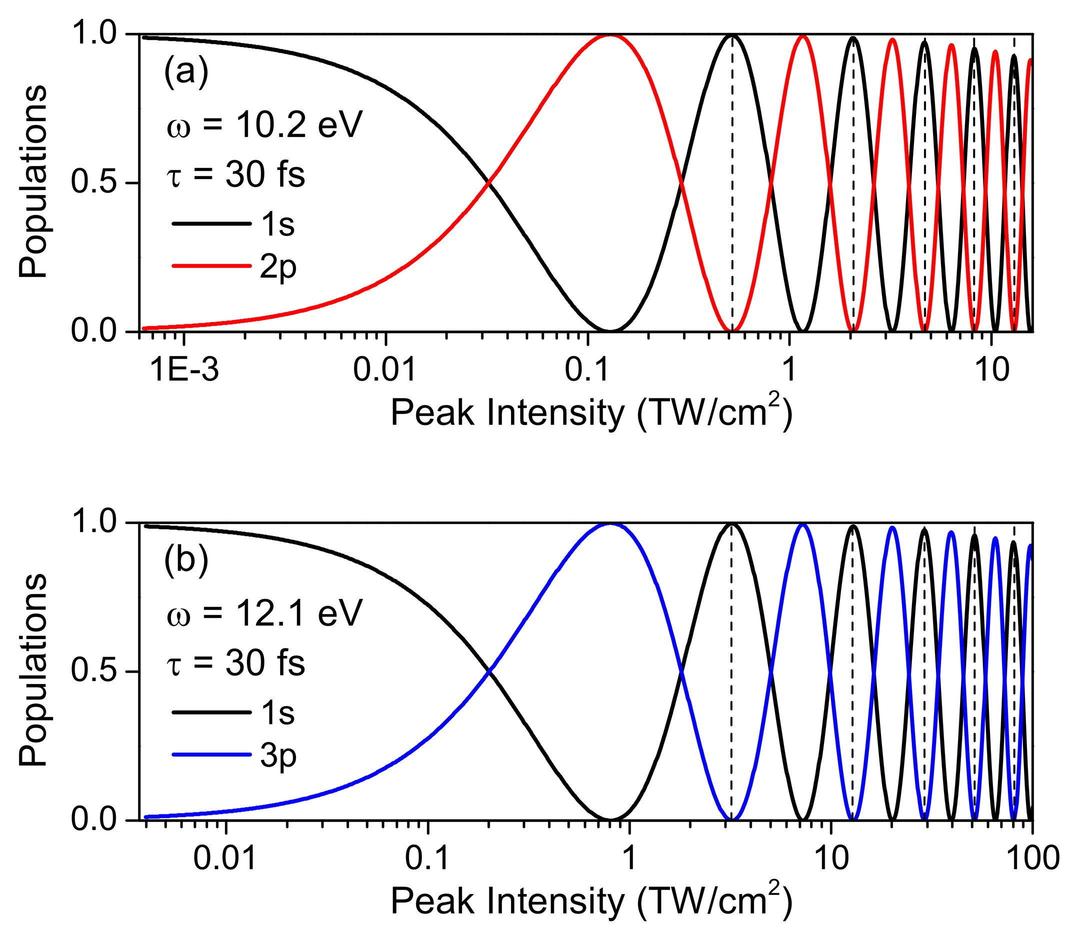

Figure 3. The populations of the ground state (1s) and the intermediate resonant state np ($n = 2, 3$) of hydrogen, after the laser pulse has expired, as functions of the peak intensity I_0. The results for the gaussian pulse of 30 fs duration and carrier frequency: (a) $\omega = 3/8\,\mathrm{a.u.} = 10.2\,\mathrm{eV}$ and (b) $\omega = 4/9\,\mathrm{a.u.} = 12.1\,\mathrm{eV}$ which fit to the energies of 1s → 2p and 1s → 3p transitions, respectively, are shown. The vertical dashed lines indicate the peak intensities at which the atom manages to complete an integer number of Rabi cycles during the pulse.

The photoelectron energy spectra shown in Fig. 4 are calculated for the peak intensities corresponding to the maxima of the ground-state population, indicated in Fig. 3 by vertical lines.

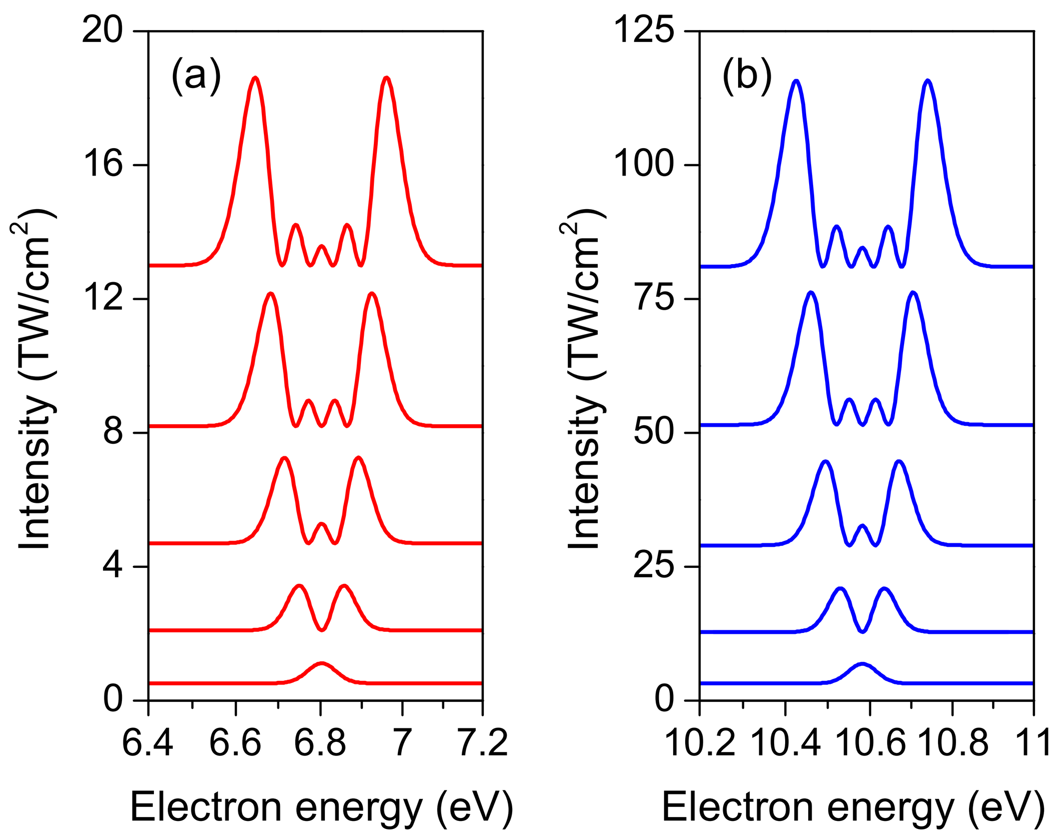

Figure 4. Photoelectron energy spectra computed for the peak intensities indicated in Fig. 3 by vertical dashed lines and two values of the carrier frequency: (a) $\omega = 10.2\,\mathrm{eV}$ and (b) $\omega = 12.1\,\mathrm{eV}$, which fit to the excitation energies of 2p and 3p state, respectively. The corresponding weak field values of the photoelectron energy are: (a) $\varepsilon_0 = 6.80\,\mathrm{eV}$ and (b) $\varepsilon_0 = 10.58\,\mathrm{eV}$.

Demekhin and Cederbaum [3] demonstrated that the modulations in the photoelectron spectra are results of *the interference of two photoelectron waves emitted with the same kinetic energy at two different times.* For each value of ε one wave is emitted at time $t_1(\varepsilon)$, when the pulse is growing, and another at time $t_2(\varepsilon)$, when it decreases. For a gaussian pulse these two times are

$$t_1(\varepsilon) = -t_2(\varepsilon) = \tau\sqrt{\ln[\Delta/(\varepsilon - \varepsilon_0)]}, \quad (10)$$

where $\Delta = D\mathcal{E}_0/2$.

References

[1] H. Friedrich, Theoretical Atomic Physics, (Springer-Verlag, Berlin Heidelberg, 2006) p. 446.

[2] P. V. Demekhin and L. S. Cederbaum, Phys. Rev. A **83**, 023422 (2011).

[3] P. V. Demekhin and L. S. Cederbaum, Phys. Rev. A **86**, 063412 (2012).