**Publ. Astron. Obs. Belgrade No. 102 (2022), 53 - 56** *Contributed Paper*

# PHOTOELECTRON ENERGY SPECTRA IN SEQUENTIAL TWO-PHOTON IONIZATION OF HYDROGEN BY GAUSSIAN AND HALF-GAUSSIAN LASER PULSES

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**Abstract.** Energy spectra of photoelectrons produced in sequential two-photon ionization of hydrogen by gaussian and half-gaussian laser pulses are studied using a three-level model (1s, 2p, continuum). The spectra show an intensity dependent splitting of the resonant peak and associated modulations. The splitting can be attributed to the existence of two dressed states whose quasi-energies repel each other by the field-induced coupling. The modulations can be explained by the interference of electron waves emitted at different times during the pulse duration.

## 1. INTRODUCTION

We study the sequential two-photon ionization of the hydrogen atom by an intense short laser pulse and analyze interference effects in the photoelectron energy spectrum (PES). The atom, which was initially in its ground state (1s), is resonantly excited into the intermediate 2p state by the absorption of a single photon of energy $\omega = E_2 - E_1 = 3/8$ a.u. and subsequently ionized by a second photon (see Fig. 1).

In order to determine the populations of atomic states during the action of the laser pulse and after, and to obtain the PES, we calculate the evolution of atomic state $|\psi(t)\rangle$ by solving the time-dependent Schrödinger equation (in atomic units)

$$i\frac{\mathrm{d}}{\mathrm{d}t}|\psi(t)\rangle = H|\psi(t)\rangle \tag{1}$$

with the initial condition $|\psi(t_0)\rangle = |1\mathrm{s}\rangle$. The total Hamiltonian has the form $H = H_0 + W$, where $H_0$ is the Hamiltonian of the field-free (bare) atom, while the term $W(t)$ describes the atom-field interaction. We consider a linearly polarized laser pulse whose electric component, directed along the z-axis, reads

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

$\mathcal{E}_0 g(t)$ is the time-dependent amplitude of the electric field strength, where $g(t)$ is the pulse envelope and $\omega$ is the carrier frequency of the pulse. Then the interaction term in the dipole approximation has the form $W(t) = z\mathcal{E}(t)$, where $z$ is the projection of the electron-nucleus distance in the field direction.

## 2. THE THREE-LEVEL MODEL

### 2. 1. EQUATIONS FOR THE AMPLITUDES FOR POPULATION OF STATES

In the case of resonant excitation of an intermediate state (here 2p), the other excited states are nonessential and at weak fields their role in the ionization process may be neglected, i.e. the process may be adequately described within the three-level

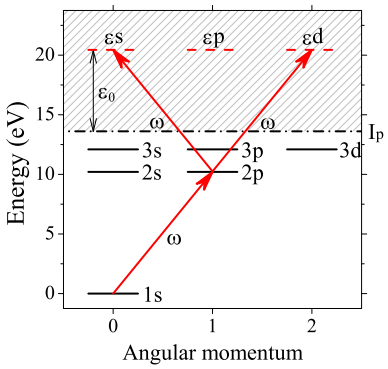


Figure 1: Energy level scheme of the hydrogen atom and the two-photon absorption paths for transitions from the ground (1s) state to the final continuum states ($\varepsilon$s and $\varepsilon$d) via one-photon resonant excitation of 2p state.

model (1s, 2p, continuum). Then, the atomic state at time $t$ reads (Demekhin and Cederbaum 2012)

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

where $a_I(t)$, $a_R(t)$ and $a_\varepsilon(t)$ are the time-dependent amplitudes for the population of states $|I\rangle \equiv |1s\rangle$ (initial), $|R\rangle \equiv |2p\rangle$ (resonant) and $|F\varepsilon\rangle$ (final), respectively. The states $|R\rangle$ and $|F\varepsilon\rangle$ have been multiplied with the phase factors $e^{-i\omega t}$ and $e^{-2i\omega t}$ in order to simplify the set of equations for the amplitudes.

By inserting Eq. (3) in the Schrödinger equation (1) and applying the rotating wave approximation (Steck 2020) and the local approximation (Demekhin and Cederbaum 2011), one obtains the following set of equations for the amplitudes (Demekhin and Cederbaum 2012)

$$\begin{aligned} i\dot{a}_I &= \frac{1}{2} D^* \mathcal{E}_0\, g(t)\, a_R(t), \\ i\dot{a}_R &= \frac{1}{2} D \mathcal{E}_0\, g(t)\, a_I(t) + \Big(E_R - \frac{i}{2}\Gamma g^2(t) - \omega\Big) a_R(t), \\ i\dot{a}_\varepsilon &= \frac{1}{2} d_\varepsilon\, \mathcal{E}_0\, g(t)\, a_R(t) + (I_p + \varepsilon - 2\omega) a_\varepsilon(t), \end{aligned} \tag{4}$$

where $D = \langle R|z|I\rangle$ and $d_\varepsilon = \langle F\varepsilon|z|R\rangle$ are the dipole transition matrix elements for the excitation of the intermediate state and for its subsequent ionization, respectively. Here we set the ground state energy to zero ($E_I = 0$, as in Fig. 1). Then the energies of the resonant and final (continuum) states are $E_R = I_p + E_2 = 3/8$ a.u. and $E_F = I_p + \varepsilon$, where $I_p = 1/2$ a.u. is the ionization potential of the hydrogen atom and $\varepsilon$ is the kinetic energy of photoelectrons. The resonant value of $\varepsilon$ is $\varepsilon_0 = 2\omega - I_p$ (see Fig. 1). Finally, $\Gamma = 2\pi\,|d_{\varepsilon_0}/\mathcal{E}_0 2|^2$ 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 $|F\varepsilon\rangle$.

### 2. 2. DRESSED STATES AND THE ENERGY SPLITTING

The resonantly coupled dynamics of states $|I\rangle$ and $|R\rangle$ in the first two of Eqs. (4) is governed by the $2 \times 2$ Hamiltonian

$$\mathcal{H} = \begin{pmatrix} 0 & \frac{1}{2}\Omega_0^* g(t) \\ \frac{1}{2}\Omega_0 g(t) & -\frac{i}{2}\Gamma g^2(t) \end{pmatrix}, \tag{5}$$

where $\Omega_0 = D\mathcal{E}_0$ is the frequency of Rabi flopping between populations of the coupled states at the peak value of laser intensity. By solving the eigenvalue problem of Hamiltonian (5) we obtain two dressed states as superpositions $|\pm\rangle \approx (|I\rangle \pm |R\rangle)/\sqrt{2}$ and the corresponding complex eigenenergies $E_\pm(t) \approx \pm\frac{1}{2}\Omega_0 g(t) - \frac{i}{4}\Gamma g^2(t)$. Due to the imaginary parts of $E_\pm$, dressed states $|\pm\rangle$ are decaying, i.e. they are two decoupled resonances. The real parts of $E_-$ and $E_+$ move apart as the pulse arrives, and towards each other as the pulse expires, estimating the splitting of the resonant peak in the PES $\Delta\varepsilon \sim \Omega_0 g_0$, where $g_0$ is the maximum value of envelope $g(t)$ (usually $g_0 = 1$).

## 3. RESULTS

The evolution of the ground state of the hydrogen atom exposed to the laser pulse of carrier frequency $\omega = E_R = 3/8$ a.u. $= 10.2$ eV has been calculated for two pulse shapes: (a) the gaussian shape $g(t) = e^{-t^2/\tau^2}$ with $\tau = 30$ fs and (b) the half-gaussian shape $g(t) = e^{-t^2/\tau^2}\mathrm{H}(t)$ with $\tau = 60$ fs ($\mathrm{H}(t)$ is the Heaviside step function). The computed dipole transition matrix elements for the excitation and ionization, used in Eqs. (4), are $D = 0.744936$ a.u. and $d_{\varepsilon_0} = 0.407759$ a.u. Figure 2 shows the evolution of populations of the ground (1s) and excited 2p state for the pulses of these two shapes and peak intensity $I_0 = 1\,\mathrm{TW/cm^2}$ ($I_0 = \mathcal{E}_0^2/(8\pi\alpha)$, $\alpha = 1/137$), while figure 3 shows the populations of these states as functions of $I_0$ in the domain of $10^9$-$10^{13}$ $\mathrm{W/cm^2}$ after the pulses have expired. One can see that the latest populations for the two pulses practically coincide.

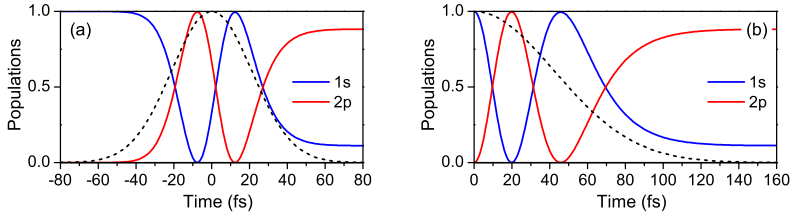


Figure 2: (a) Populations of the ground (1s) and the excited 2p state, calculated as $|a_I(t)|^2$ and $|a_R(t)|^2$, respectively, at the sequential two-photon ionization of hydrogen by a gaussian laser pulse of $1\,\mathrm{TW/cm^2}$ peak intensity, $\tau = 30$ fs and the carrier frequency $\omega = 10.2$ eV which fits to the energy of $1\mathrm{s} \to 2\mathrm{p}$ transition. (b) The populations obtained using the half-gaussian pulse with $\tau = 60$ fs and the same frequency and intensity. The dashed lines represent the envelopes of the laser pulses.

Figure 4 shows the photoelectron energy spectra calculated for the two shapes of the laser pulse and the peak intensities $I_0$ marked in figure 3 by vertical lines. For each value of $I_0$ the spectra consist of the resonant peak whose splitting, according to relation $\Delta\varepsilon \sim \Omega_0 g_0$, increases with the peak value of field strength $\mathcal{E}_0$ ($\sim \sqrt{I_0}$).

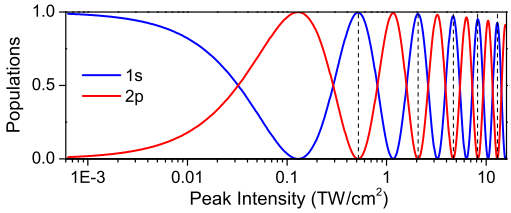


Figure 3: Populations of the ground (1s) and excited 2p state of hydrogen as functions of the laser peak intensity after the laser pulse has expired. The results obtained for the gaussian and half-gaussian pulses of the same carrier frequency $\omega = 10.2\,\mathrm{eV}$ with $\tau = 30\,\mathrm{fs}$ and $\tau = 60\,\mathrm{fs}$, respectively, which practically coincide, are presented. The vertical dashed lines indicate the peak intensities at which the atom manages to complete an integer number of Rabi cycles during the pulse.

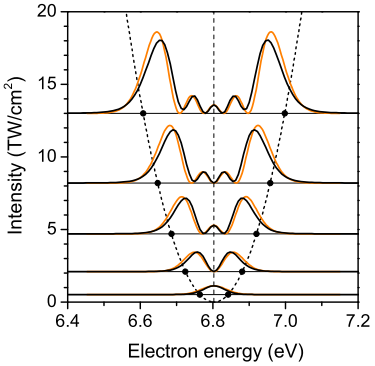


Figure 4: Photoelectron energy spectra represented by distributions $w(\varepsilon) = |a_\varepsilon(3\tau)|^2$ calculated for the gaussian and half-gaussian laser pulses (orange/black lines) with the peak intensities marked in Fig. 3 by vertical lines. Black dots mark the real parts of $E_\pm(0)$, whose separation ($\approx \Omega_0$) estimates the splitting of the resonant peak.

Demekhin and Cederbaum (2012) analyzed the modulations in the PES obtained for the photoionization with the gaussian pulse. They explained the occurrence of modulations between the positions of $E_\pm$ resonances by the interference of two photoelectron waves emitted with the same kinetic energy at two different times – at time when the pulse is growing and at time when it decreases. Our calculations, however, show that similar modulations exist also in the case of photoionization with the half-gaussian pulse, that has no growing part. Based on this, we conclude that the modulations are due to the interference of electron waves emitted all the time during the pulse duration, rather than at two specific times.

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