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Multiple ionization of atoms with xuv attosecond pulses: two-photon double ionization of helium with 50 eV photons

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Abstract. We consider two-photon double ionization of helium by two xuv photons in the region around the sequential ionization threshold. We show that, on the attosecond timescale, the mechanism for double ionization is dominated by the absorption of one photon by each electron in the fundamental state He($1s^2$). We examine the dynamics of two-photon double ionization of helium for an averaged photon energy ω =50 eV, with a pulse duration of two optical cycles. The double ionization rate, energy and angular distributions are calculated by solving the time-dependent Schrödinger equation. Results are discussed on the basis of a model.

1. Introduction

The study of correlated multi-electron dynamics in atoms and molecules is a necessary step towards a better understanding of multi-electron ejection in complex systems interacting with intense lasers. In the xuv domain two-electron ejection *via* one-photon absorption has received considerable attention from both theoretical and experimental sides. This process is now well understood. In the non-linear interaction regime where more than one photon is absorbed, multi-electron ejection has been studied in the infrared frequency domain with intense fields. In this case, experimental evidence as well as theoretical analysis have favored a physical mechanism based on a rescattering picture for non-sequential double ionization [1]. Double ionization of helium by several XUV photons has a much lower probability to occur, and therefore requires xuv sources that are more intense than the conventional ones. The only available sources are high order harmonic generation (HOHG) and free electron lasers (FEL). Two-photon double ionization (TPDI) of helium has been measured using both types of sources. However, the level of uncertainty in the data is very high [2] and therefore precludes a reliable comparison with the large number of existing theoretical results. On the theoretical side, the TPDI has indeed received considerable attention (see [3] for a list of references, up to 2008), but there are large

discrepancies between the results for the total and differential TPDI cross sections obtained with different models in the direct regime ($\omega < 2$ a.u.) [3]. Therefore TPDI is still clearly an open problem which may require considerable effort from both theorists and experimentalists to solve.

Over the last years we have thoroughly studied TPDI in both direct ($\omega < 2$ a.u.) and sequential ($\omega > 2$ a.u.) regimes. In particular, we investigated the dynamics of TPDI in case of sub-fs pulses by studying both the electron angular and energy distributions [4]. We have shown that the electrons are predominantly emitted back-to-back along the polarization axis in direct TPDI, revealing the important role of dynamical screening [3]. For sub-fs pulses, sequential TPDI tends to a transient regime and back-to-back emission becomes the dominant process [5], as in the direct regime. Here we investigate TPDI of helium with 50 eV photons in the limit of attosecond (as) pulses. With the help of a simple model we show that TDPI is dominated by the absorption of one photon by each electron of the initial state. Atomic units are used throughout this paper, unless stated otherwise.

2. Theoretical approach

2.1. Time-dependent perturbative and non-perturbative treatments

Our theoretical approach is based on the solution of the time-dependent Schrödinger equation (TDSE)

$$i\frac{\partial}{\partial t}\Psi(\vec{r}_1,\vec{r}_2,t) = \left[-\frac{1}{2}\nabla^2_{r_1} - \frac{1}{2}\nabla^2_{r_2} - \frac{2}{r_1} - \frac{2}{r_2} + \frac{1}{r_{12}} + D_G(t)\right]\Psi(\vec{r}_1,\vec{r}_2,t), \tag{1}$$

where r_1 and r_2 are the radial coordinates of both electrons and $r_{12} = |\vec{r}_1 - \vec{r}_2|$ is the inter-electronic distance. $D_G(t)$ describes the dipole interaction of the system with the oscillating field either in the length gauge ($G \equiv L$) or in the velocity gauge ($G \equiv V$):

$$D_L(t) = \vec{E}(t)\cdot(\vec{r}_1 + \vec{r}_2), \tag{2}$$

$$D_V(t) = -i\vec{A}(t)\cdot(\vec{\bigtriangledown}_1 + \vec{\bigtriangledown}_2). \tag{3}$$

$\vec{A}(t)$ denotes the vector potential and $\vec{E}(t) = \hat{z}E_0 f(t)\sin(\omega t+\phi)$ the electric field which oscillates at the frequency ω with a phase ϕ and which is assumed linearly polarized along the z-axis. Here $f(t)$ is the pulse envelope given by:

$$f(t) = \begin{cases} \cos^2(\pi t/T);\ |t| \le \dfrac{T}{2}, \\ 0;\ |t| > \dfrac{T}{2}. \end{cases} \tag{4}$$

In this expression, T represents the total duration of the pulse. We have:

$$\vec{A}(T/2) = -\int_{-T/2}^{T/2} dt\vec{E}(t) = 0, \tag{5}$$

for any pulse duration and phase. This means that even for few-cycle pulses there is no static field component, and problems related to gauge invariance [6] do not arise.

Our method to solve the TDSE has been described in detail in [7, 8]. Briefly, we first expand the full wave packet of the system $\Psi(\vec{r}_1,\vec{r}_2,t)$ in terms of its field-free eigenstates. The wave functions associated with these eigenstates are calculated within a spectral method that consists of diagonalizing the atomic Hamiltonian in a basis of products of one-electron square integrable functions of the radial coordinates r_1 and r_2 and bipolar harmonics of the electron

angular coordinates. The square integrable functions are either Coulomb Sturmian functions or B-splines.

After propagating the initial wave packet until such time that the interaction with the pulse has died out, we are left with the task of calculating the ionization probabilities, a task which poses a challenge in all theoretical approaches. We calculate the double ionization probability using two different approaches. The first one is a simple projection of the wave packet onto a product of Coulomb functions, at $t \geq T/2$. The second one, developed in the context of Sturmian functions, uses the Jacobi-matrix method [10] to generate in the Coulomb Sturmian basis, a multichannel scattering wave function that describes accurately the single continuum of He while incorporating the correct asymptotic conditions. Projecting the final wave packet $\Psi(\vec{r}_1, \vec{r}_2, t)$ at the end of the pulse on this function provides a tool to disentangle the single- and double-ionization components [8].

In order to probe how electron correlations in the ground state of He affect the double-escape process, we have developed a simple model in which electron correlations enter only in the ground state of He. This model is based on lowest order time-dependent perturbation theory. This is justified since in the high frequency and low intensity regimes considered here the ponderomotive shift of the electrons is negligible compared to the photon energy. For two-photon double ionization, the probability amplitude reads:

$$U^{(2)} = -\sum_{\alpha} \langle \Psi_f | z_1 + z_2 | \Psi_\alpha \rangle \langle \Psi_\alpha | z_1 + z_2 | \Psi_i \rangle K(E_0, \omega, \phi, E_i, E_\alpha, E_f), \tag{6}$$

where the function K is given by:

$$\begin{aligned} K(E_0, \omega, \phi, E_i, E_\alpha, E_f) &= \int_{-T/2}^{T/2} d\tau_1 E_0 f(\tau_1) \sin(\omega\tau_1 + \phi) e^{i\omega_{f\alpha}\tau_1} \\ &\quad \int_{-T/2}^{\tau_1} d\tau_2 E_0 f(\tau_2) \sin(\omega\tau_2 + \phi) e^{i\omega_{\alpha i}\tau_2}. \end{aligned} \tag{7}$$

$\omega_{f\alpha} = E_f - E_\alpha$ and $\omega_{\alpha i} = E_\alpha - E_i$ with E_i, E_α and E_f the initial, the intermediate, and the final state energy respectively. E_0 is the maximum field amplitude. The wave function of the initial state Ψ_i is written as follows:

$$\Psi_i(\vec{r}_1, \vec{r}_2) = \sum_{l,\nu,n} \phi^{l,l}_{\nu,n} \mathcal{A} F^{l,l}_{\nu,n}(r_1, r_2) \Lambda^{0,0}_{l,l}(\Omega_1, \Omega_2), \tag{8}$$

where the coefficients $\phi^{l,l}_{\nu,n}$ are calculated by diagonalizing the atomic Hamiltonian of He. $\mathcal{A}$ is the antisymmetrization operator and $\Lambda^{0,0}_{l,l}(\Omega_1, \Omega_2)$ is a bipolar harmonic in the angular coordinates of both electrons. The radial function $F^{l,l}_{\nu,n}(r_1, r_2)$ is written as a product of two hydrogenic bound states of principal quantum number ν and n respectively:

$$F^{l,l}_{\nu,n}(r_1, r_2) = \psi_{\nu,l}(r_1)\psi_{n,l}(r_2). \tag{9}$$

Let us stress that the above expression (8) for Ψ_i is not correct because this expansion does not contain continuum states. However, it contains some radial and angular correlation. In this calculation of $\Psi_i(\vec{r}_1, \vec{r}_2)$, we have included 4 pairs (ℓ, ℓ) of the electron angular momenta ((0,0), (1,1), (2,2) and (3,3)) and taken both ν and n varying from $1+\ell$ to $4+\ell$. The resulting estimate of the ground state energy is -2.84 a.u. The intermediate states Ψ_α and the final state Ψ_f are defined as an antisymmetrized product of bound or/and Coulomb states. We have tested this model by comparing the results with those obtained by the approach described at the beginning of this section. The agreement is good both qualitatively and quantitatively [5]. In addition,

this model also gives reasonable results in the direct regime.

Before discussing the TDSE results we briefly present several models which include electron correlations in various degrees of approximation. Their pertinence is discussed with respect to the pulse duration; they illustrate the strong dependence of the TPDI dynamics on T.

2.2. The zero-order approximation and the independent electron model

We recall first a previous study where the role of electron correlation was treated in lowest order perturbation theory [9]. In this approach, the He ground state is described by the $1s^2$ configuration, i.e., by a product of two hydrogenic orbitals $1s$. The other states are the single continuum state He(1skp) and double continuum He(kpk'p), also represented by hydrogenic orbitals (all orbitals are calculated with $Z = 2$). In this approximation the energy $E^0_{1s^2}$ of the ground state would then be given by $2E^0_{1s} + < 1s^2|1/r_{12}|1s^2 >$ where E^0_{1s} is the hydrogenic energy of $1s$ and $< 1s^2|1/r_{12}|1s^2 >$ the electron interaction energy. For long pulse durations, it is possible to derive an analytical expression for the photoelectron energy spectrum, it reads

$$|U_{k,k'}(T \to \infty)|^2 \propto \frac{|\langle kp|z_1|1s\rangle\langle k'p|z_2|1s\rangle|^2}{[(E_k + E_{k'} - E^0_{1s^2} - 2\omega)^2 + \frac{1}{4}\gamma^2_{He}][(E_k - E^0_{1s} - \omega)^2 + \frac{1}{4}\gamma^2_{He^+}]}, \tag{10}$$

where γ_{He} and γ_{He^+} are the photoionization widths of He and He^+, respectively. $\langle kp|\mu|1s\rangle$ represents the dipole coupling between the $1s$ orbital and the kp continuum. E_k and $E_{k'}$ are the energies of the ejected electrons. The electron spectrum shows two peaks separated by the energy $E_I =< 1s^2|1/r_{12}|1s^2 >$. E_I is the electron interaction energy exchanged during the ionization process. Here it is worth noticing that the effective widths of the peaks is the result not of a single Lorentzian but of a convolution.

If now we fully neglect the electron interactions, i.e. if we adopt the independent electron model, it is easy to show that the expression (6) splits into the product of two terms representing the absorption of one photon from each $1s$ orbital. $U^{(2)}$ is written

$$U^{(2)} = -\sqrt{2}\langle kp|z_1|1s\rangle K_M(E_{1s}, E_k)\langle k'p|z_2|1s\rangle K_M(E_{1s}, E_{k'}), \tag{11}$$

where

$$K_M(E_i, E_f) = \int_{-T/2}^{T/2} d\tau E_0 f(\tau) sin(\omega\tau + \phi) e^{i(E_f - E_i)\tau}. \tag{12}$$

It is worth noticing that the final state has a unique angular configuration of the electron pair — it is (1,1). The corresponding angular distribution peaks when both electrons move in the same direction with the same energy, a feature which can occur in the absence of electron correlation [3].

2.3. A simple model to investigate TPDI with ultrashort pulses

We return now to equation (6). Our aim is to develop a simple model to describe TPDI in the limit of ultrashort pulses. The initial state is now represented by a simple product of 1s orbitals with a screening charge of 27/16. For ultrashort pulses (e.g., two optical cycles with a photon energy of 50 eV), the function $K(E_0, \omega, \phi, E_i, E_\alpha, E_f)$ shows a slow dependence on the intermediate energy E_α. Therefore, we replace the value E_α by the resonant energy $E_r = E_i + \omega$. It is straightforward to show that

$$U^{(2)} = -\langle\Psi_f|(z_1 + z_2)^2|\Psi_i\rangle K(E_0, \omega, \phi, E_i, E_r, E_f). \tag{13}$$

Here it is interesting to note that the matrix element $\langle\Psi_f|(z_1 + z_2)^2|\Psi_i\rangle$ exhibits two types of couplings. The first one, $z_1 z_2$, is associated with the absorption of one photon by each

electron in Ψ_i, as illustrated in equations (10) and (11). The second term, z_i^2 (with $i = 1, 2$), represents the absorption of two photons by one electron, the second electron being ionized through electron interactions. This latter term does not contribute when the electron interactions are fully neglected. These two terms have a crucial impact on angular distributions in the final state; this is discussed in the following section.

3. Results and discussion

We consider TPDI with 50 eV photons and a pulse duration of two optical cycles (2c), the intensity is 10^{13} W/cm^2. Note that one optical cycle (1c) has a duration of 82 as. We have checked that three photon absorption is negligible at the intensity considered.

First we have compared our model (see section 2.3) with TDSE calculations; the agreement is excellent. We have checked that the coupling $2z_1z_2$ dominates over z_1^2 and z_2^2 (although the latter are non-negligible).

L	(l_1, l_2)	$0c$	$2c$	$4c$
2	(1,1)	$0.18\ 10^{-8}$	$0.17\ 10^{-8}$	$0.16\ 10^{-8}$
0	(1,1)	$0.82\ 10^{-9}$	$0.60\ 10^{-9}$	$0.48\ 10^{-9}$
2	(0,2)	$0.71\ 10^{-10}$	$0.24\ 10^{-9}$	$0.34\ 10^{-9}$
0	(0,0)	$0.49\ 10^{-10}$	$0.17\ 10^{-9}$	$0.24\ 10^{-9}$

Table 1. Dominant angular pair contributions in double continuum produced by two-photon absorption at $t = T/2$ (i.e., at the end of the pulse), $t = T/2 + 2c$ and $t = T/2 + 4c$. The laser parameters are the same than in figure 1.

After solving the TDSE, angular distributions have been extracted by projecting the total wavefunction $\Psi(\vec{r}_1, \vec{r}_2, t)$ (see equation 1) onto an antisymmetrized product of Coulomb functions (see equations (12) and (13) in [5]). Using the latter functions, calculated with $Z = 2$, we neglect the electron interactions in double continuum but these interactions are included in TDSE and free propagation. Table 1 shows the dominant angular pair contributions calculated at different times after the end of the pulse. The table clearly shows that the pair $(1, 1)$ dominates at $t = T/2$ (i.e., at the end of the pulse). We see that the contributions of other angular pairs, in particular (0,2), increases at $t = T/2 + 2c$ and $t = T/2 + 4c$. Here it is worth mentioning that when a three particle system disintegrates the term 'angular momentum distribution' becomes meaningless. Indeed, it is in principle necessary to take into account more and more angular momenta as the particles separate further, the only exception being the collinear configuration [11].

Figure 1 shows the angular distribution in the case were both electrons share a similar energy (0.4 a.u.). The first electron is emitted along the z axis with an angle of $\theta = 0$. As in the precedent case the information is extracted from $\Psi(\vec{r}_1, \vec{r}_2, t)$ at various times t. At $t = T/2$ the figure shows that the second electron is preferentially emitted in the opposite direction ($\theta = \pi$) but there is a non-negligible probability that both electrons are emitted in the same direction. At the end of the short pulse both electrons are close to the nucleus and strongly interact. After four cycles (i.e., at $t = T/2 + 4c$) the latter contribution is negligible, the distribution clearly shows that the electron are emitted in opposite directions. The results agree with the analysis of table 1, in fact, the presence of the single angular pair (1,1) in the double continuum would lead to a symmetric distribution [3].

In conclusion, for a pulse with attosecond duration and photon energy 50 eV the double

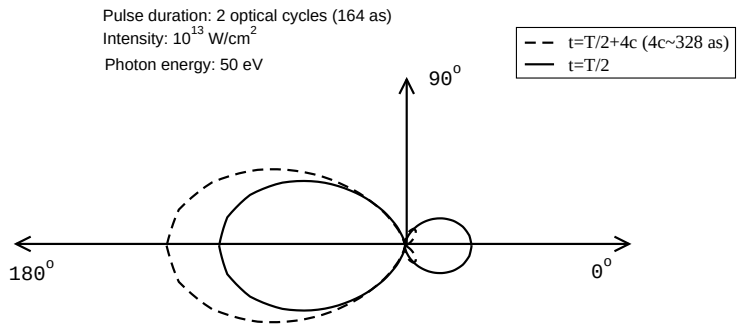

Figure 1. TPDI angular distribution, in polar coordinates. Both electrons have an energy of 0.4 a.u., one of them being emitted along the z axis ($\theta = 0$). The calculations are performed at the time $T/2$ (i.e., at the end of the pulse, full line) and at $T/2 + 4c$ (i.e., four cycles after the end of the pulse, dashed line). The laser parameters are indicated in the figure.

ionization process is dominated by the absorption of one photon by each electron. This leads, at the end of the pulse, to a wave packet with strongly interacting electrons where the double continuum is dominated by an angular pair $(1, 1)$. Subsequently, the strong electron interaction in the double ionization channel generates other angular pairs, resulting physically in electron emission in opposite directions. Therefore, in the limit of ultrashort pulse durations, it is possible to disentangle the effect of the laser coupling and electron correlations. In the attosecond regime the angular and energy distributions show unexpected features; they will be analyzed in details in a forthcoming publication.

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