

Probing electron-electron correlation with attosecond pulses

B. Piraux^{1,a}, J. Bauer², S. Laulan³, and H. Bachau³

¹ Laboratoire de Physique Atomique, Moléculaire et Optique, Université catholique de Louvain, 2 chemin du cyclotron, 1348 Louvain-la-Neuve, Belgium

² Katedra Fizyki Jądrowej i Bezpieczeństwa Radiacyjnego, Uniwersytet Łódzki, ul. Pomorska 149/153, 90-236 Łódź, Poland

³ Centre Lasers Intenses et Applications^b, Université de Bordeaux I, 351 cours de la Libération, 33405 Talence, France

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Abstract. We study two-photon double ionization of helium in its ground state at sufficiently low laser intensities so that three and more photon absorptions are negligible. In the regime where sequential ionization dominates, the two-photon double ionization one-electron energy spectrum exhibits a well defined double peak structure directly related to the electron-electron correlation in the ground state. We demonstrate that when helium is exposed to subfemtosecond or attosecond pulses, both peaks move and their displacement is a signature of the time needed by the He^+ orbital to relax after the ejection of the first electron. This result rests on the numerical solution of the corresponding non-relativistic time-dependent Schrödinger equation.

PACS. 32.80.Rm Multiphoton ionization and excitation to highly excited states (e.g., Rydberg states) – 32.80.Dz Autoionization

1 Introduction

The direct exploration of the bound electron dynamics in atoms or molecules requires XUV pulses as short as a few hundreds of attoseconds (as). Over the past decade, various schemes aimed at generating as-pulse trains [1–6] and single as-pulses [7,8] have been studied. Among these schemes, the most promising one towards the production of as-pulses is based on high order harmonic generation with few-cycle Ti:S laser pulses in noble gas [9,10]. The first convincing experimental evidences [11,12] of the production of as-pulses have been obtained recently. At present, substantial efforts are made in two directions: (i) a complete characterization of an as-pulse and (ii) an increase of the XUV fluence available from the high-harmonic sources. These efforts should pave the route towards time-resolved spectroscopy in the attosecond domain.

In this contribution, we consider the helium atom and demonstrate that it is possible to probe with one XUV as-pulse, the electron-electron correlation in its ground state. We study the two-photon double ionization (DI) (2γ , $2e$) process from $\text{He}(1^1\text{S})$ at sufficiently low laser intensities so that three and more photon absorptions are negligible. As shown in a previous work [13] and by contrast to one-photon double ionization, this process is dominated by transition channels that require no

interaction between the electrons. Furthermore, for photon energies ω larger than 2 a.u. and long pulse durations, the (2γ , $2e$) process is mainly sequential while for $\omega < 2$ a.u. ($39.5 \text{ eV} < \hbar\omega < 54.4 \text{ eV}$) it becomes direct. For $\omega > 2$ a.u., the one-electron energy spectrum exhibits two peaks which are actually spaced by the correlation energy E_c in the ground state. This correlation energy is defined here by the difference between the “exact” ground state energy $E_{\text{He}(1^1\text{S})}$ and the ground state energy E_{1s^2} obtained within the independent model. Note that if both electrons were completely independent (electron-electron correlation neglected in the ground state), the one-electron energy spectrum would exhibit only one peak. To the correlation energy, we associate a correlation time defined in a.u. by $1/E_c$. The question that is addressed here is the following: how is the one-electron energy spectrum modified when the interaction time becomes of the order of the correlation time for $\omega > 2$ a.u.? We show that together with an expected broadening of the two peaks, these peaks move towards each other until we reach a situation in which both peaks merge: in that case, the interaction time is so short that it is impossible to distinguish between the direct and the sequential process. The displacement of the peaks is actually a direct signature of the time the He^+ orbital takes to relax after the ejection of the first electron. These effects have been first discussed in the context of (2γ , $2e$) processes in the helium isoelectronic series [14]. When the photon energy is high enough so that autoionizing states may play a role, we show that

^a e-mail: Piraux@fyam.ucl.ac.be

^b UMR 5107 du CNRS

the (2γ , $2e$) process is still strongly dominated by the sequential process in agreement with the recent results of Nikolopoulos *et al.* [15].

Our results for the one-electron energy spectrum are obtained by solving numerically the (non-relativistic) time-dependent Schrödinger equation (TDSE). We use a spectral method of configuration interaction type. Products of L^2 integrable functions are used to represent the radial part of the total wavefunction. The L^2 integrable functions are either Coulomb Sturmian functions [16] or B-splines [17]. Owing to the complexity of the calculations, we systematically compare the results obtained with the two types of basis functions. In the following, atomic units are used throughout unless otherwise stated.

2 Theoretical approach

2.1 Atomic structure calculation

Let us first briefly describe our theoretical approach. A given helium wavefunction Ψ_α of energy E_α is solution of the following equation:

$$H\Psi_\alpha = E_\alpha\Psi_\alpha, \quad (1)$$

where H is the non-relativistic Hamiltonian; it writes:

$$H = -\frac{1}{2}\Delta_1 - \frac{1}{2}\Delta_2 - \frac{Z}{r_1} - \frac{Z}{r_2} + \frac{1}{|\mathbf{r}_1 - \mathbf{r}_2|}. \quad (2)$$

Z denotes the nucleus charge and \mathbf{r}_1 and \mathbf{r}_2 are the radial coordinates of the two electrons. For a given total angular momentum L and its projection M , we expand the solution of equation (1) as follows:

$$\Psi_\alpha^{L,M}(\mathbf{r}_1, \mathbf{r}_2) = \sum_{\lambda,l,\nu,n} c_{\alpha,\nu,n}^{\lambda,l,L,M} \mathcal{A} \times \frac{F_{\lambda,\nu}(r_1)}{r_1} \frac{F_{l,n}(r_2)}{r_2} A_{\lambda,l}^{L,M}(\Omega_1, \Omega_2), \quad (3)$$

where \mathcal{A} is the antisymmetrisation operator and $c_{\alpha,\nu,n}^{\lambda,l,L,M}$ the expansion coefficient. λ and l are the angular momenta of the electrons and $A_{\lambda,l}^{L,M}$ a bipolar spherical harmonics function of Ω_1 and Ω_2 , the angular coordinates of both electrons. The functions $F_{l,n}$ are L^2 integrable functions which are either Coulomb Sturmian functions or B-splines. For a given angular momentum l , the Coulomb Sturmian functions $S_{l,n}^k(r)$ are defined as follows:

$$S_{l,n}^k(r) = N_{l,n}^k r^{l+1} L_{n-l-1}^{2l+1}(2kr), \quad (4)$$

where $N_{l,n}^k$ is a normalization constant and L_{n-l-1}^{2l+1} a Laguerre polynomial. The wavevector k plays the role of a scaling factor and the index n varies between $l+1$ and $N+l+1$ where N is the number of Coulomb Sturmian functions for a given l . Note that these functions form a complete and discrete basis and are solution of the radial hydrogenic Sturm-Liouville eigenvalue problem [18]. The

B-spline functions $B_n^k(r)$ of order k are piecewise polynomials of degree $k-1$. The index n varies between 1 and N_b where N_b is the number of B-splines. The N_b B-spline functions are spanned, along the radial axis, in a ‘‘box’’ defined from $r=0$ to R_{\max} . The B-spline sequence is chosen so that $B_1^k(0) = B_{N_b}^k(R_{\max}) = 0$ in order to fulfil the correct boundary conditions within the box.

A direct diagonalization of equation (1) gives the eigenenergies E_α as well as the expansion coefficients $c_{\alpha,\nu,n}^{\lambda,l,L,M}$ of the bound and continuum states. The autoionization width of the doubly excited states can be obtained by means of a complex rotation of the Hamiltonian H . Both bases lead to an accurate description of the ground state of helium as well as many singly and doubly excited states. It is not the purpose of this contribution to analyse further the adequacy of both bases in the present context. This has already been discussed elsewhere [14]. In fact, it turns out that these bases are very efficient in many situations in atomic and molecular physics, see for instance [19–23] for a more detailed discussion.

2.2 Time-dependent calculations

We now consider the interaction between the atom and the laser field. Within the dipole approximation, the time-dependent Schrödinger equation to be solved is given in the velocity gauge, by:

$$i\frac{\partial}{\partial t}\Psi_v(\mathbf{r}_1, \mathbf{r}_2, t) = \left[H + \sum_{i=1,2} \mathbf{A}(t) \cdot \mathbf{p}_i \right] \Psi_v(\mathbf{r}_1, \mathbf{r}_2, t). \quad (5)$$

The vector potential, polarized along the z -axis, is defined in the interval $[-T/2, T/2]$ as:

$$\mathbf{A}(t) = A_0 \left(\cos \frac{\pi}{T} t \right)^2 \cos(\omega t) \mathbf{e}_z, \quad (6)$$

where T is the total pulse duration and ω the photon energy. Similarly, in the length gauge, the time-dependent Schrödinger equation reads:

$$i\frac{\partial}{\partial t}\Psi_l(\mathbf{r}_1, \mathbf{r}_2, t) = \left[H + \sum_{i=1,2} \mathbf{E}(t) \cdot \mathbf{r}_i \right] \Psi_l(\mathbf{r}_1, \mathbf{r}_2, t), \quad (7)$$

with the electric field defined as:

$$\mathbf{E}(t) = -\frac{\partial}{\partial t} \mathbf{A}(t). \quad (8)$$

The total time-dependent wavefunction is expanded in the basis of the field-free atomic eigenstates (see Eqs. (1–3)), normalized to unity:

$$\Psi_{v,l}(\mathbf{r}_1, \mathbf{r}_2, t) = \sum_{\alpha,L} C_{\alpha,L}^{v,l}(t) \Psi_\alpha^{L,M}(\mathbf{r}_1, \mathbf{r}_2). \quad (9)$$

Both equation (5) and equation (7) are integrated over the total pulse duration T . This integration is carried out

within the interaction picture with a usual Runge-Kutta type formula. Note that by working in the interaction picture, we eliminate all the oscillations of the total wavefunction that are driven by its field-free evolution. The initial condition is given by:

$$\begin{aligned}\Psi_l(\mathbf{r}_1, \mathbf{r}_2, t = -T/2) &= \Psi_v(\mathbf{r}_1, \mathbf{r}_2, t = -T/2) \\ &= \Psi_{in}^{L=0, M=0}(\mathbf{r}_1, \mathbf{r}_2),\end{aligned}\quad (10)$$

where $\Psi_{in}^{L=0, M=0}(\mathbf{r}_1, \mathbf{r}_2)$ is the initial state of the field-free system *i.e.* the ground state of helium in the present case. In the following it is assumed that $M = 0$.

At the end of the pulse, the population of a given stationary state Ψ_α is obtained by projecting this state on the total wavefunction. Our finite basis however describes the continuum states over finite distances. It is therefore very difficult to separate single and double ionization contributions from the final wavefunction. In addition to the fact that the two-electron single and double continuum states may be degenerate in energy, the *numerically built* positive energy states contain necessarily both single and double continuum components since our finite basis does not allow to describe either the single or the double continuum wavefunction in the asymptotic region. In the present case, the energy distribution of the two ejected electrons is obtained by projecting the final state wavefunction (in the length or the velocity gauge) on an uncorrelated product of two Coulomb wavefunctions $\Psi_{c,p}$:

$$\begin{aligned}\frac{\partial^2 P_{DI}}{\partial \epsilon_1 \partial \epsilon_2} &= \rho(\epsilon_1) \rho(\epsilon_2) \int d\Omega_1 \int d\Omega_2 \\ &\times |\langle \Psi_{c,p_1} \Psi_{c,p_2} | \Psi_{v,l}(t = T/2) \rangle|^2,\end{aligned}\quad (11)$$

where $\rho(\epsilon)$ is the density of states in the continuum. Whether or not the above expression is a good approximation is still an open question. The results which will be presented in the following indicate however that correlation in the final state do not play a crucial role. On the other hand, the implementation of a new algorithm taking into account the electron correlation (at all orders) in the final state is under way. In the case of B-splines, it is possible to built up a double continuum wavefunction following a method described by Nikolopoulos *et al.* [24]. In the case of the Coulomb Sturmian functions, we use the J -matrix method [25,26] to generate an accurate single continuum wavefunction. The double continuum component of the final wavefunction (after the elimination of the negative total energy component) is then obtained by subtracting from this function, its projection on the accurate single continua. Switching to momentum space allows to extract the energy distribution of the ejected electrons. The one-electron energy spectrum in the double continuum is obtained by integrating expression (11) over one of the electron energies. An additional integration over the energy of this electron provides the total double ionization probability P_{DI} .

All the results presented here have been obtained by means of two bases (a B-spline and a Sturmian basis). In our B-spline basis, the number of B-splines per electron

Table 1. Angular terms for the B-spline basis.

Angular momentum L	(λ, l) angular pairs	Number of combination terms
$L = 0$	(0, 0) (1, 1) (2, 2) (3, 3)	5100
$L = 1$	(0, 1) (1, 2)	5000
$L = 2$	(0, 2) (1, 1) (1, 3)	6275
$L = 3$	(0, 3) (1, 2)	5000

Table 2. Angular terms for the Sturmian basis.

Angular momentum L	(λ, l) angular pairs	Number of combination terms
$L = 0$	(0, 0) (1, 1) (2, 2) (3, 3)	5100
$L = 1$	(0, 1) (1, 2) (2, 3)	7500
$L = 2$	(0, 2) (1, 1) (1, 3) (2, 2)	7095

angular momentum $N_b = 50$. This means that 2500 combination terms are used for each pair of electron angular momenta (λ, l) when $\lambda \neq l$ and 1275 when $\lambda = l$. The box radius $R_{\max} = 50$ a.u. and the order k of the B-splines is equal to 7. A linear knot sequence is used. The angular terms $((\lambda, l)$ pairs) used in the calculations are given in Table 1. In the case of the Sturmian basis, we also use 50 Sturmian functions per (λ, l) pair except for the (2, 2) pair ($L = 2$) where only 40 functions were included. In Table 2, we give the angular terms that have been used in the calculations. The non-linear parameter k is equal to 2 for $L = 0$, 1 for $L = 1$ and 0.7 for $L = 2$. The fact that the number of total angular momenta is limited to 2 or 3 is justified because the laser intensities considered here are relatively low (of the order of 10^{14} watt/cm²) well within the validity of the lowest order of perturbation theory. The number of pairs of electron angular momenta (λ, l) per L is also limited. The inclusion of further pairs increases significantly the size of each L block of the atomic Hamiltonian matrix making the diagonalization problem almost computationally untractable. However, this is not crucial in the present case because the atomic process that is analyzed here is dominated by interaction channels where correlation do not play any role. This means that the pairs (0, 0), (1, 1) and (0, 2) are the dominant ones in the final states. This point has been checked carefully by considering the contribution to the final cross-section of each individual pair separately. We have also checked that our results are gauge independent in both bases. It is important to note that although the time propagation is performed within the interaction picture, the calculation of the observables at the end of the pulse is carried out within the Schrödinger picture. In principle the results do not depend on the picture in which the total wavefunction is expressed. In practice however, the basis is truncated; this leads in some cases to significant differences (up to 50%) between the results obtained in both pictures. Our method has been tested for one-photon double ionization. We reproduce with a good accuracy the one-photon double ionization rates obtained by Shakeshaft *et al.* [27,28]. The difference with our data is of the order of 5%.

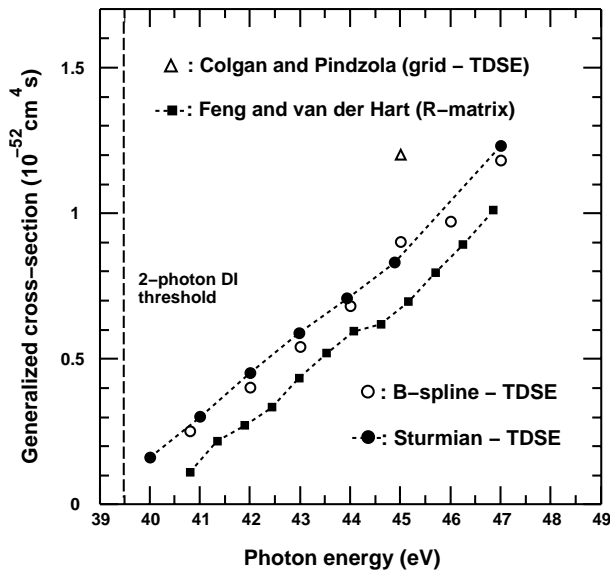


Fig. 1. Generalized cross-section for two-photon double ionization of the ground state of helium. The present results (open and full circle) have been obtained by solving the TDSE using B-splines (open circle) and Sturmian functions (full circle). These results are compared to those of Colgan and Pindzola [29] and Feng and van der Hart [30]. The position of the double ionization threshold is also indicated.

3 Two-photon double-ionization

3.1 Generalized cross-section

For photon energies below 2.2 a.u. (≈ 60 eV) *i.e.* outside the regime of the autoionizing resonances, it is meaningful to define the two-photon double ionization generalized cross-section $\sigma(2\gamma, 2e)$. By assuming that the adiabatic approximation is valid and that the pulse shape is given by equation (6), $\sigma(2\gamma, 2e)$ expressed in cm^4s , is given by:

$$\sigma(2\gamma, 2e) = 2.268 \times 10^{-23} \times \frac{\omega^3 \times P_{\text{DI}}}{I_0^2 \times T}, \quad (12)$$

where ω is in eV, and the peak intensity I_0 in watt/cm^2 . The total duration T is expressed in number of optical cycles. Since the generalized cross-section is actually defined within the lowest order of perturbation theory, we use a sufficiently low peak intensity $I_0 = 10^{14}$ watt/cm^2 . In the present case, the total duration of the pulse $T = 10$ optical cycles. Our results obtained with both bases (as described in the previous section) are presented in Figure 1. They are compared to the results obtained by Colgan and Pindzola [29] and Feng and van der Hart [30]. Colgan and Pindzola use a time-dependent approach similar to our method but in which the total wavefunction is built up on a grid. Feng and van der Hart use a *R*-matrix formalism taking into account the electron-electron correlation in the final state. We are not showing here the results obtained within the lowest order of perturbation theory [31,32] which are 5 times higher than the results presented here. As a matter of fact, there is no clear *consensus* so far regarding which

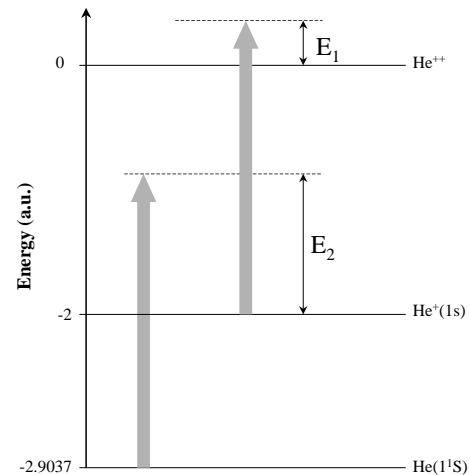


Fig. 2. Schematic diagram of the levels involved in the two-photon double electron ejection.

are the most accurate calculations. However, we note the striking agreement between the results obtained with each of our two bases [33] and the fact that our data are very close to those of Feng and van der Hart. This last point seems to indicate that at least in this case, the inclusion of electron-electron correlation in the double continuum is not crucial.

3.2 One-electron energy spectrum

Let us now study in detail the one-electron energy spectrum. As shown in a previous work [13], $(2\gamma, 2e)$ processes are dominated by transition channels that do not require any electron-electron correlation. At this stage, it is convenient to consider two frequency regimes. For frequencies larger than 2 a.u., the process is sequential and proceeds as shown in Figure 2: after the absorption of one photon and the emission of the first electron, the system is in the ground state of He^+ . It then absorbs another photon which ejects the second electron. We have found that the probability for the system to reach the double continuum through any channel involving an excited state of He^+ is negligible. This means that if the photon frequency is less than 2 a.u., the process is direct. For $\omega > 2$ a.u., the one-electron energy spectrum exhibits two peaks, one at $E_1 = E_{\text{He}^+(1s)} + \omega$ and the second one at $E_2 = E_{\text{He}(1^1S)} + \omega - E_{\text{He}^+(1s)}$ (see Fig. 2). The distance between these two peaks is given by: $E_c = E_2 - E_1 = E_{\text{He}(1^1S)} - 2E_{\text{He}^+(1s)} = E_{\text{He}(1^1S)} - E_{1s^2}$. E_c is actually the ground state correlation energy defined here as the difference between the “exact” ground state energy $E_{\text{He}(1^1S)} = -2.9037$ a.u. and the ground state energy $E_{1s^2} = -4$ a.u. obtained within the independent model. If both electrons were completely independent, the one-electron energy spectrum would exhibit only one peak at an energy of $\omega - 2$ in a.u. In the present case, the correlation energy is 1.1 a.u. corresponding to a correlation time $\tau_c = 1/E_c = 0.9$ a.u. or 21.8 attoseconds (as). In the

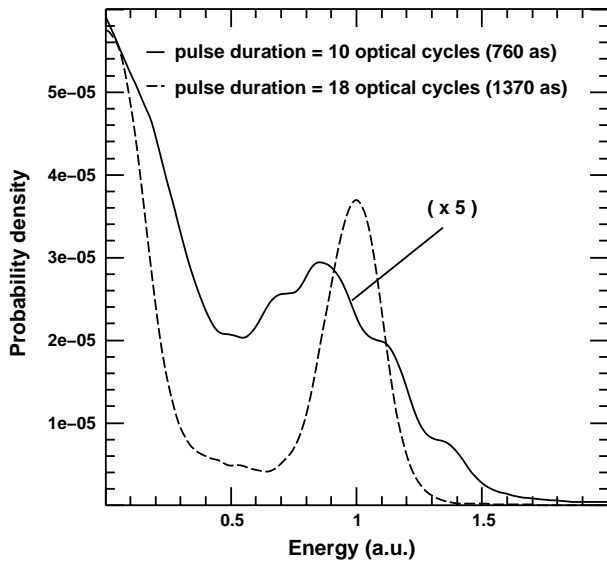


Fig. 3. One-electron energy spectrum resulting from the double ionization of He(1^1S) with an ultrashort pulse whose peak intensity $I_0 = 10^{14}$ watt/cm 2 and $\omega = 2$ a.u. Two pulse durations are considered: 10 optical cycles (≈ 760 as) and 18 optical cycles (≈ 1370 as). For clarity, the data corresponding to the full line have been multiplied by a factor 5.

following, we study how the one-electron energy spectrum modifies for ultrashort pulses.

In Figure 3, we show the one-electron energy spectrum for a frequency $\omega = 2$ a.u. and a peak intensity $I_0 = 10^{14}$ watt/cm 2 . Two pulse durations are considered: 10 optical cycles (≈ 760 as) and 18 optical cycles (≈ 1370 as). The results shown here have been obtained by using a Sturmian basis. Similar data have been produced with the B-spline basis. We first note that $\omega = 2$ a.u. is actually the threshold frequency for the sequential process to occur. It is the reason why the position of the low energy peak is not clearly defined. For decreasing pulse durations, the peaks broaden as expected and start to move. We clearly see that the high energy peak is shifted towards lower energies by about 0.15 a.u. or 4.1 eV. Note that the width of the peaks stays much smaller than the correlation energy. We show in Figure 4 the position in eV of the high energy peak as a function of half the duration of the pulse for the same frequency and peak intensity. When the full pulse duration decreases from 18 to 6 a.u., the high energy peak moves from 27.5 eV to 19 eV. This significant shift is interpreted as follows.

For very short pulses (of the order of the correlation time τ_c), the first electron ejection occurs necessarily over an extremely short time leaving the other electron in a non-stationary state *i.e.* a superposition of many states of He $^+$. In presence of the field, this non-stationary state can relax towards the ground state of He $^+$ unless the interaction time with the pulse is shorter than the relaxation time. If this is the case, it becomes meaningless to distinguish between the direct and the sequential process. Since the intermediate state is non-stationary, its energy is not defined. Its averaged energy however will be higher

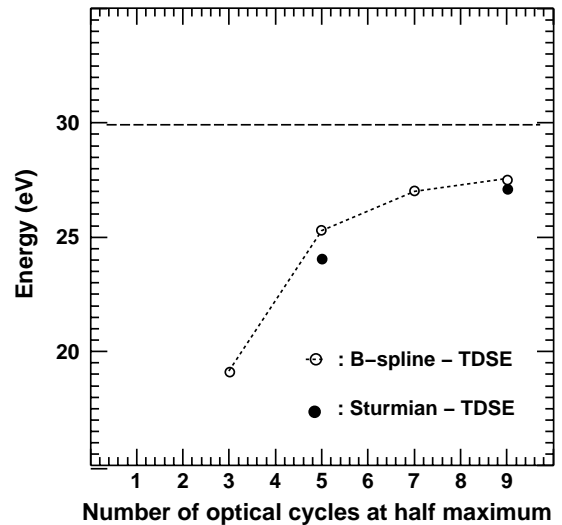


Fig. 4. Position of the most energetic peak as a function of half the duration of the pulse in optical cycle for the same case as in Figure 3. The results obtained with both bases are compared and the position of the most energetic peak in the limit of long pulses (dashed line) is also indicated.

than $E_{\text{He}^+(1s)}$; this explains why both peaks are moving towards each other. This effect takes place already for pulse durations much longer than the correlation time τ_c . This results from the fact that double ionization occurs only in a very small time interval around the maximum of the pulse. In the present case and for a pulse duration of 18 optical cycles, we found that the total double ionization probability is 24×10^{-6} . Note that perturbation theory is clearly valid in the present conditions.

In the following we consider two other frequencies: one lower than 2 a.u. where the (2γ , $2e$) process is direct and another one larger than 2 a.u. where doubly excited states of helium might play a role as intermediate states. In Figure 5, we show the energy distribution of both electrons for a frequency $\omega = 1.65$ a.u., a peak intensity $I_0 = 10^{14}$ watt/cm 2 and a pulse duration of 10 optical cycles. The calculations have been performed with a Sturmian basis. We clearly see that the distribution is rather flat by contrast to a sequential process which according to the previous discussion is characterized by two sharp peaks at positions symmetric with respect to the straight line $E_1 = E_2$. When the frequency is such that doubly excited states could play a role as intermediate states, it is legitimate to ask whether or not the (2γ , $2e$) process is still dominated by transition channels that require no electron-electron correlation. Note that for helium and for subfemtosecond pulses, most of the doubly excited states can be considered as bound states. In order to answer to the above question, we consider $\omega = 2.34$ a.u. With this frequency and a long pulse duration, the first photon absorption would drive the atom almost on resonance with the third doubly excited P-state above the first single ionization threshold. We show in Figure 6 the one-electron energy spectrum for a peak intensity $I_0 = 5 \times 10^{14}$ watt/cm 2 and two

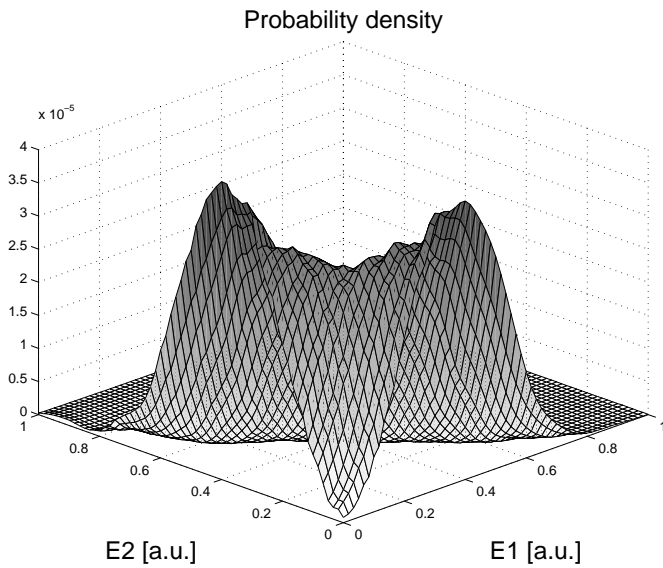


Fig. 5. Electron energy distribution for $\omega = 1.65$ a.u., $I_0 = 10^{14}$ watt/cm² and a pulse duration of 10 optical cycles.

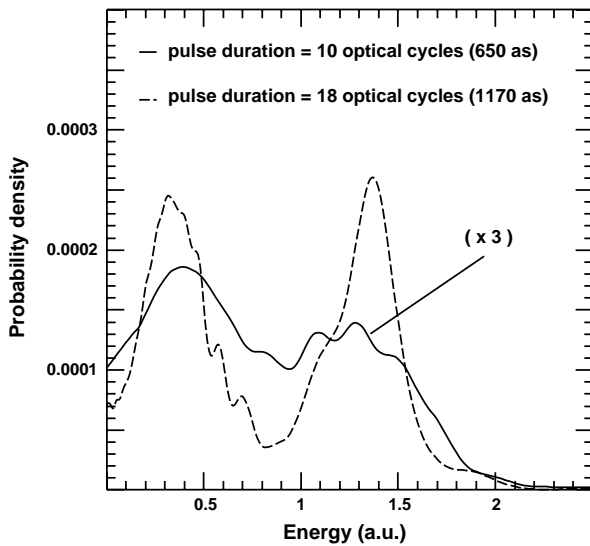


Fig. 6. One-electron energy spectrum resulting from the double ionization of He(1¹S) with an ultrashort pulse whose peak intensity $I_0 = 5 \times 10^{14}$ watt/cm² and $\omega = 2.34$ a.u. Two pulse durations are considered: 10 optical cycles (≈ 650 as) and 18 optical cycles (≈ 1170 as). For clarity, the data corresponding to the full line have been multiplied by a factor 3.

pulse durations: 10 optical cycles (650 as) and 18 optical cycles (1170 as). The data have been obtained with a Sturmian basis. We have checked that the B-spline basis produces similar data. The one-electron energy spectrum exhibits two well defined peaks around $E_1 = 0.3$ a.u. and $E_2 = 1.35$ a.u. for the 18 optical cycle pulse. These peak positions are precisely those expected on the basis of the above discussion regarding the sequential process. Furthermore, we see that the peaks move towards each other when the pulse duration is decreased. Therefore, it is clear in agreement with previous results [15] that in these con-

ditions of frequency, the $(2\gamma, 2e)$ process is still essentially sequential and that the electron-electron correlations do not play any role.

4 Conclusion

We have studied the two-photon double ionization of He in its ground state by attosecond pulses and focused our attention on the one-electron energy spectrum. For frequencies equal or larger than 2 a.u., the ionization process is sequential and the one-electron energy spectrum exhibits two well defined peaks spaced by the electron correlation energy in the ground state of He. For ultrashort pulse durations in the subfemtosecond regime, we showed that the peaks move towards each other for decreasing pulse durations; the distance between the peaks is directly related to the relaxation time of the second electron orbital after the ejection of the first one. All the results presented in this contribution have been obtained by solving numerically the time-dependent Schrödinger equation.

Taking into account the constant progress regarding the production and characterization of attosecond pulses, we hope that an experiment will be performed soon. In this experiment, both the ion and one of the electrons must be detected in coincidence.

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