Dynamics of the direct double ionization of helium
by two XUV photons

Bernard Piraux¹, Emmanuel Foumouo¹, Philippe Antoine¹
and Henri Bachau²
¹ Laboratoire de Physique Atomique, Moléculaire et Optique (unité PAMO), Université Catholique de Louvain, 2, chemin du Cyclotron, B-1348 Louvain-la-Neuve, Belgium
² Centre des Lasers Intenses et Applications (CELIA), Université Bordeaux I-CNRS-CEA, 351, Cours de la Libération, F-33405 Talence, France
E-mail: bernard.piraux@uclouvain.be

Abstract. As a highly correlated process, direct two-photon double ionization of helium represents a very challenging theoretical problem. A wide range of computational methods have been used to evaluate generalized cross sections. However, despite considerable efforts, a quantitative agreement between all calculations has not been reached. In this contribution, we address several issues regarding the origin of the discrepancies in the total cross section and the behaviour of this cross section close to the sequential regime. Our \textit{ab initio} calculations are based on the numerical solution of the time-dependent Schrödinger equation by means of a spectral method combined with Jacobi-matrix calculations. This method which, in principle, takes fully into account electron correlations, provides also a consistent physical picture of the double escape mechanism.

1. Introduction

The study of the dynamics of the double ionization of atoms poses real difficulties mostly because of the long range nature of the Coulomb interactions between the electrons. Until recently, the main efforts have focused on one-photon double ionization of helium. This process has served as a testing ground for understanding the subtle role of the electron correlations in the double electron escape. The agreement of the predictions of most of the theoretical approaches (see [1] and references therein) with the (absolute) experimental data [2, 3] obtained with synchrotron radiation, have led to a clear picture of the double escape mechanism.

Moving from single photon processes to two-photon processes leads to a rich variety of new phenomena enabling a deeper exploration of electronic correlations. But, it also introduces serious theoretical and experimental complications. From the experimental point of view, the main problem is the following. Two-photon double ionization (TPDI) of atoms is a second order process which has a very low probability to occur. Thereby, it requires XUV sources that are more intense than the conventional ones. At the present stage, the only available sources are high order harmonic generation (HOHG) and the free electron laser (FEL). However, the characterization of the pulses (temporal and spatial profile of the beam, ...) emitted by these two new sources is very difficult to obtain and even not well known in some cases. It is therefore realistic to say that at the present stage, an absolute measurement of the TPDI generalized
cross section is clearly out of reach. From the theoretical point of view, the calculation of the TPDI generalized cross section based on the lowest non-vanishing order of time-independent perturbation theory is extremely difficult. This calculation involves an infinite summation on all intermediate stationary P-states of helium as well as the knowledge of the final double continuum state whose asymptotic behaviour is unknown. An alternative way of evaluating the total TPDI generalized cross section consists in solving the time-dependent Schrödinger equation to obtain the final two-electron wavepacket at the end of the interaction with the pulse. Although, this method is relatively easier to implement, it suffers the same problem of the evaluation of the final state wavefunction. All approaches are therefore based on some approximations, that have to be properly understood and controlled. Note that the problem of the corrections to perturbation theory is not an issue here, taking into account the pulse durations and the intensities provided by the new FEL and HOHG sources.

Owing to the complexity of the evaluation of the TPDI generalized cross section, it is not surprising that there is presently no consensus among all theoretical calculations. In this contribution, we discuss this issue and analyse the behaviour of the TPDI generalized cross section as a function of the photon energy. This analysis is performed by means of our time-dependent approach that is based on a spectral method combined with Jacobi-matrix calculations. Besides, our method provides a consistent physical picture of the double escape mechanism.

This contribution is organized as follows. We start by defining three distinct regimes of TPDI as a function of the photon energy and the pulse duration. We then describe briefly our time-dependent method and compare our results for the total TPDI generalized cross section with the others obtained with different approaches. The last section is devoted to the double escape mechanism.

2. TPDI regimes

For sufficiently long pulses and photon energies higher than 2 a.u. (the ionization potential of He$^+$ in its ground state), the TPDI is predominantly sequential: the absorption of a photon by one of the electron is followed by the relaxation of the residual He$^+$ ion in its ground state and subsequently, by the absorption of a photon by the second electron. In these conditions, the process is dominated by transition channels that require no interaction between the electrons [4]. This leads to an electron energy distribution that exhibits two sharp peaks, one associated with the single ionization of He and the other one to the ionization of He$^+$. These two peaks, are in fact separated by what we call the dielectronic interaction energy $E_{di}$. It actually represents the energy difference between the true ground state energy of He and the independent-particle energy of this ground state. Note that in a sequential process, it is the first ejected electron that carries this dielectronic interaction energy $E_{di}$. For sufficiently low intensities (i.e. in the absence of significant depletion of the initial state), the probability for sequential TPDI is proportional to the square of the pulse duration. This means that in this sequential regime, the notion of (generalized) cross section is meaningless. In fact, a proper quantitative description of the sequential TPDI process requires the solution of two rate equations [5].

For sufficiently long pulses and photon energies higher than 1.45 a.u. (TPDI energy threshold), the process can be direct. In that case, the absorption of the two photons occurs within a time less than the relaxation time of the He$^+$ ion. In the range of photon energies from 1.45 a.u. to 2 a.u., the double ionization process is purely direct because the sequential TPDI process is energetically forbidden. It is precisely this range of photon energies that is considered in this contribution. For sufficiently low intensities, the probability of TPDI is now proportional to the pulse duration. Note that in the direct regime, both ejected electrons share the dielectronic interaction energy $E_{di}$. 
There is a third TPDI regime called transient that corresponds to pulse durations which are of the order or less than the relaxation time of the ion He \(^+\) [6]. This relaxation time is defined as \(2\pi/E_{di}\). It is of the order of 140 attosecond for He in its ground state. Note that for such ultrashort pulses, the notion of photon looses its meaning. It is also important to stress that in this regime, the TPDI process is neither purely direct nor sequential.

3. Theoretical approach

The theoretical approach we use to evaluate the total TPDI generalized cross section is described in detail in [1]. Here, we only give an outline of the method. It rests on the solution of the time-dependent Schrödinger equation (TDSE). We work in the dipole approximation and perform the calculations both in the length and the velocity gauges. We use a sine square pulse whose total duration is about 10 optical cycles. One of the drawbacks of the time-dependent methods is the impossibility of separating the direct and sequential processes. In the direct regime considered here (photon energies between 1.45 a.u. and 2 a.u.), three-photon sequential processes occur while leading to double ionisation of He. It is therefore important to work at peak intensities (here, about \(10^{12}\) Watt/cm\(^2\)) where these third order sequential processes can be neglected. Furthermore, two-photon sequential processes may also occur close to the sequential regime as a result of the finite bandwidth of the pulse. When the photon energy gets too close to 2 a.u., it is therefore necessary to significantly increase the duration of the pulse.

The TDSE is solved by means of a spectral method of configuration interaction type which consists in expanding the atomic state wavefunctions in a finite basis of products of Coulomb-sturmian functions of the electron radial coordinates and bipolar harmonics of the angular coordinates. The time propagation of the two-electron wavepacket is carried out in the atomic basis and within the interaction picture by means of an explicit Runge-Kutta scheme. Once the total wavefunction is obtained at the end of the interaction with the pulse, we first subtract from it all bound state contributions in order to get the totally ionized wavepacket. We then have to project this wavepacket on the double continuum wavefunctions in order to extract the information about double ionization. At this stage, it is important to make the following remarks. The double continuum should be described by a multichannel scattering wavefunction. The coupling between all channels is the result of the dielectronic Coulomb interaction term. The exact asymptotic behaviour of this function is not known. Finally, since our finite basis does not describe the asymptotic region, the states of positive energy (usually called pseudostates) obtained by diagonalizing the atomic Hamiltonian contain necessarily both single and double continuum components that cannot be disentangled. In order to overcome these difficulties, we find more appropriate to generate the single continuum multichannel scattering wavefunctions. By projecting the totally ionized wavepacket on these functions and summing on all single ionization channels, we get the total probability of single ionization. Subtracting this single ionization probability from the total ionization probability provides the probability of double ionization.

To generate an accurate single continuum multichannel scattering wavefunction, we use the Jacobi matrix method which has been introduced by Heller and Yamani [7]. This method which is also of spectral type, bears close resemblance to the R-matrix theory. The configuration space is divided into two regions. In the inner region, the space is spanned by the same finite sturmian basis used to build the atomic eigenstates. In the outer or asymptotic region, it is assumed that the outgoing electron moves in a screened Coulomb potential. In order to reproduce the correct asymptotic behaviour of the outgoing electron wavefunction in each channel, it is necessary, at least implicitly, to expand this wavefunction in the complete sturmian basis therefore implying to take into account an infinite number of Coulomb sturmian functions. Within this approach however, the double continuum channels are described in an approximate way. Asymptotically,
one of the electrons is described by a Coulomb wave while the other one (the inner one) is described by a pseudostate. Let us note that within the convergent close-coupling (CCC) method, this approximation is made in whole space [8].

In order to assess the accuracy of our approach, we studied various one-photon processes where very accurate benchmark results and in some cases, absolute experimental data exist. In particular, we considered the single ionization (with or without excitation of the residual ion) of He and $\text{H}^-$ for photon energies below and above the double ionization threshold as well as the double ionization of He and $\text{H}^-$. In all cases, our results are in very good agreement with the other data even close to the thresholds (see [1] for more details). It is worth mentioning that the case of one-photon double ionization of He is a stringent test because the double ionization probability (which is obtained by subtracting the single ionization probability from the total ionization probability) is two orders of magnitude smaller than the probability of single escape.

4. TPDI total cross section

Our results for the (generalized) TPDI total cross section are shown in Fig. 1 where they are compared to other calculations and to the first experimental results. The noticeable discrepancies between the data clearly illustrate the difficulty of treating TPDI to high accuracy. Before discussing the theoretical results in detail, let us comment first on the two experimental data. The open black circle with error bar is the experimental result of Sorokin et al [9] at 42.8 eV photon energy. They used the FEL at Hamburg (FLASH) and estimated the TPDI cross section from a measurement of the He$^{++}$ to He$^+$ yield ratio. It is worth noting that their estimation depends on the spatial and temporal profiles chosen for the analysis of their experimental data. In fact, the cross section has to be multiplied by a factor $2\sqrt{2}$ if Gaussian spatial and temporal profiles are used instead of square profiles. The bottom of the vertical line at 40.8 eV photon energy gives a realistic estimation of a lower bound of the TPDI cross section obtained from a new experiment performed by the group of Midorikawa [10] with HOHG. In this new experiment, they measured the He$^{++}$ to He$^+$ yield ratio and made a systematic study of the possible saturation effects affecting the production of He$^+$. We now turn to the theoretical data. There are clearly two important issues to discuss: the origin of the important discrepancies between the results and the behaviour of the cross section close to the sequential limit for a photon energy of $2 \text{ a.u. (54.4 eV)}$. As we mentioned above, there are two types of theoretical approaches: the time-independent and the time-dependent ones. The time-independent approaches include the method of Horner et al [11] based on the Floquet theory combined with an exterior complex scaling (ECS) technique, the method of Ivanov and Kheifets [12] who use the lowest order perturbation theory with a CCC final state, the flux formula of Shakeshaft [13] and the R-matrix Floquet calculations of Feng and van der Hart [14]. Note that, according to their authors, the results of the last two methods are not fully converged with respect to the size of the basis. The time-dependent methods include the multichannel theory of Nikolopoulos and Lambropoulos [15] and the method of Feist et al [16] which consists in propagating the full electron wavepacket on a grid and in projecting this wavepacket onto a product of two Coulomb functions (with effective charge equal to 2) a long time after the end of the interaction with the pulse. By means of a time-propagation method of spectral type, Laulan and Bachau [17] make a similar calculation, but the projection onto the product of two Coulomb functions is performed at the end of the interaction with the pulse. We also perform such calculation with our spectral method (curve labelled TDSE-NC in Fig. 1) in addition to our Jacobi matrix calculations (curve labelled TDSE-FC in Fig. 1). All time-dependent approaches differ by the way electron-electron correlations are treated in the final state.

In order to gain more insight in this problem, we analyse in Fig. 2 what kind of electron
correlation in the final state is responsible for the strong enhancement of the cross section. It is important to stress that neglecting completely the electron correlations in the single continuum wavefunction generated by the Jacobi matrix method leads as expected, to a cross section that is in perfect agreement with what is obtained by projecting directly the final wavepacket onto a product of two Coulomb functions. If on the other hand, we only take into account the radial correlations in the calculation of the single continuum wavefunction with our Jacobi matrix method, we obtain a cross section that is very close to our fully correlated result. It is therefore the radial coupling between the different channel components of the single continuum wavefunction that is responsible for the strong effect we observe in the TPDI cross section. This conclusion gives support to the TPDI mechanism which is discussed in the last section of this contribution. It is worth noting that in the sequential regime, i.e. for photon energies higher than the last section of this contribution, it is worth noting that in the sequential regime, i.e. for photon energies higher than

**Figure 1.** TPDI total cross section as a function of the photon energy. The continuous grey line with open circles (curve labelled TDSE-NC) are our results obtained by neglecting the final state-electron correlation and the continuous grey line with full circles (curve labelled TDSE-FC) are our results obtained by means of the Jacobi matrix method. Our results are compared to the results of Nikolopoulos and Lambropoulos (dashed line with open grey circles) [15], Ivanov and Kheifets (dashed line with full black circles) [12], Shakeshaft (continuous grey line with open triangles) [13], Laulan and Bachau (continuous line with small full grey circles) [17], Feist et al (continuous black line with large open circles) [16], Feng and van der Hart (continuous grey line with open squares) [14] and Horner et al (continuous grey line with full triangles) [11]. At 42.8 eV, the open black circle with error bar is the experimental data of Sorokin et al [9] while the bottom of the vertical black line at 40.8 eV photon energy, indicates an estimation of a lower bound of the TPDI cross section from a new experiment of Midorikawa et al [10].
Figure 2. TPDI total cross section as a function of the photon energy. The continuous grey line with full circles is our Jacobi matrix result. The dashed grey line with full triangles is our result obtained by taking only into account the radial correlations in the single continuum wavefunction generated by the Jacobi matrix method. The continuous dark line with full circles is our result obtained by projecting the final wavepacket on a product of two Coulomb functions of effective charge 2. The squares are obtained by switching off the Coulomb repulsion between the electrons in the single continuum wavefunction generated by the Jacobi matrix method.

than 2 a.u., the TPDI probabilities evaluated by means of our Jacobi matrix approach and by projecting the electron wavepacket directly onto two Coulomb functions are in agreement. This is consistent with the fact that sequential processes occur without electron correlation.

Feist et al [16] claim that the inclusion of electron correlations in the final double continuum state can be bypassed by waiting long enough after the end of the pulse before performing the projection onto a product of two Coulomb functions. It is worth noting that when they use the same sine square pulse, they obtain a TPDI total cross section that is in perfect agreement (see Fig. 8b of reference [16]) with the results of Laulan and Bachau as well as with our results in which we neglect the final state electron correlations. This does not prove that the final state electron correlations do not play a significant role. Instead, the adequacy of such description of the final state electron correlations remains an open question. The product of two Coulomb functions with effective charges 2 is not orthogonal to the single continuum eigenstates of He. Founouo et al [20] have shown that this lack of orthogonality leads to spurious effects in
the electron angular distributions, in particular, when one of the electrons is not ejected along the polarization axis. In addition, this product of two Coulomb functions has not the correct asymptotic behaviour. As shown by Madsen et al [18], because the three coulombic interactions have the same strength and equal range, the well known Brauner Briggs Klar (BBK) double continuum wavefunction [19] should be used instead of a product of two Coulomb functions. To conclude this short discussion, let us mention that important discrepancies also exist between the results of the time-independent approaches (see Fig. 1). The reasons of these discrepancies are not understood at the present stage.

The second issue is the sharp rise of the TPDI total cross section very close to the sequential limit, i.e. for photon energies close to 2 a.u.. Horner et al [11] are the first who observe the existence of this behaviour of the cross section. Later on, Feist et al [16] and Ivanov and Kheifets [12] obtained similar results. In the context of a time-dependent approach, the sharp rise of the TPDI cross section close to the sequential limit is most likely due to the sequential TPDI [5]. The frequency bandwidth of the pulse and its duration have two competing effects. On the one hand, a large frequency bandwidth increases the overlap with the sequential domain of photon energies. On the other hand, the probability for sequential TPDI varies quadratically (instead of linearly for direct transitions) with the pulse duration. The fact that Feist et al observe a much sharper rise of the TPDI cross section with longer pulses indicates that the duration of the pulse has the major effect. In addition, the fact that the TPDI cross section depends on the pulse duration also indicates that what is calculated does not represent a cross section. For that reason, we have never calculated a TPDI cross section very close to the sequential limit. The presence of this sharp rise of the TPDI cross section in the time-independent approaches is more intriguing. Horner et al explain this behaviour of the cross section by invoking the notion of ”virtual sequential ionization” [21]. Another tentative explanation is the following: to get converged results, Horner et al have to introduce a small imaginary part to the field frequency. This is equivalent to introduce artificially a pulse bandwidth therefore explaining the rise of the cross section. The reason of the sharp rise of the TPDI cross section in the calculations of Ivanov and Kheifets is not clear and certainly requires further investigations.

5. TPDI mechanism

From the above discussion, it becomes clear that electron correlations in the final state play a crucial role. We have recently investigated this issue in more details by analyzing both the singly differential cross sections (electron energy distributions) and the triply differential cross sections (electron angular distributions for a given excess energy partitioning) [20]. It turns out that angular correlations strongly favour back-to-back electron emission along the polarization axis, while dynamical screening (i.e. radial correlations) leads to an equipartition of the electron energy for photon energies ranging from the double ionization threshold to about 48 eV. Close to the double ionization threshold, each electron has to maximally screen the other electron in order to overcome the Coulomb attraction of the nucleus [22]. The fact that the screening is dynamic means that both electrons exchange energy during the double escape process. Once they are roughly at equal distance from the nucleus and have the same energy, which is the most favorable configuration for double escape to occur, the screening becomes static; the electrons do not need to exchange energy any more. A direct signature of the dynamical screening is found in the time evolution of the radial doubly ionized two-electron wavepacket. Unfortunately, the corresponding graphs cannot be reproduced here because of the lack of space. When the photon energy increases and gets larger than 48 eV, the dynamical screening is much less crucial. This manifests itself by a flat and even a U-shaped energy distribution around and above 50 eV photon energy. In order to validate this mechanism, we proposed to measure the distribution of the recoil momentum $P$ of the doubly charged ion. According to the above mechanism, this
distribution should exhibit a pronounced maximum around $P = 0$.

Acknowledgments

The authors wish to thank Laurence Malegat, Peter Lambropoulos and Robin Shakeshaft for very stimulating discussions. HB thanks the Laboratoire de Physique Atomique, Moléculaire et Optique (unité PAMO) of the Université Catholique de Louvain, for hospitality and financial support. The present work is supported by the FNRS (Fonds National de la Recherche Scientifique) through the FRFC (Fonds de la Recherche Fondamentale et Collective) program (project no. 2. 4592.07). The authors thank the Université Catholique de Louvain for providing them with an access to the supercomputer of the CISM (Calcul Intensif et Stockage de Masse) which is supported by the FNRS through the FRFC project no. 2.4556.99, "Simulations Numériques et Traitement des Données".

References

[1] Foumouo E, Lagmago Kumta G, Edah G and Piraux B 2006 Phys. Rev. A74 063409
[2] Samson J A R, Stolte W C, He Z X, Cutler J N, Lu Y and Bartlett R J (1998) Phys. Rev A57 1906
[3] Bräuning H et al. 1998 J. Phys. B 31 5149
[4] Bachau H, Lambropoulos P 1991 Phys. Rev. A44 R9
[5] Lambropoulos P (2008) private communication
[6] Foumouo E, Antoine Ph, Bachau H and Piraux B 2008 NJP 10 025017
[7] Heller E J, Yamani H A 1974 Phys. Rev. A9 1201
[8] Kheifets A S and Ivanov I A 2006 J. Phys. B 39 1731
[9] Sorokin A A, Wellhöfer M, Bobashev S V, Tiedtke K and Richter M 2007 Phys. Rev. A75 051402
[10] Antoine Ph, Foumouo E, Piraux B, Shimizu T, Hasegawa H, Nabelewski Y and Midorikawa K 2008 To be published in Phys. Rev. A
[11] Horner D A, Morales F, Recsino T N, Martin and McCurdy C W 2007 Phys. Rev. A76 030701(R)
[12] Ivanov I A and Kheifets A S 2008 J. Phys. B41 095002
[13] Shakeshaft R 2007 Phys. Rev. A76 063405
[14] Feng L and van der Hart H W 2003 J. Phys. B36 L1
[15] Nikolopoulos I A A and Lambropoulos P 2007 J. Phys. B40 1347
[16] Feist J, Nagele S, Pazourek R, Persson E, Schneider B I, Collins L A and Burgdörfer 2008 Phys. Rev. A77 043420
[17] Laulan S and Bachau H 2003 Proc. Int. Conf. on Electron and Photon Impact Ionization and Related Topics, Metz, France (IOP Conf. Ser. No. 172 ed Ancarani L U p 109
[18] Madsen L B, Nikolopoulos L A A, Kjeldsen T K and Fernández J 2007 Phys. Rev. A76 063407
[19] Brauner M, Briggs J S and Klar H 1989 J. Phys. B22 2265
[20] Foumouo E, Antoine Ph, Piraux B, Malegat L, Bachau H and Shakeshaft R 2008 J. Phys. B41 051001
[21] Horner A D, Rescino T N and McCurdy C W 2008 Phys. Rev. A77 030703(R)
[22] Rau A R P 1971 Phys. Rev. A4 207