Angular distributions in double ionization of helium under XUV sub-femtosecond radiation

P Lambropoulos\textsuperscript{1,2,4} and L A A Nikolopoulos\textsuperscript{3}

\textsuperscript{1} Institute of Electronic Structure and Laser—FORTH, PO Box 1527, GR-711 10 Heraklion (Crete), Greece
\textsuperscript{2} Department of Physics, University of Crete, PO Box 2208, GR-71003 Heraklion (Crete), Greece
\textsuperscript{3} Department of Applied Mathematics and Theoretical Physics, The Queen’s University of Belfast, Belfast BT7 1NN, UK
E-mail: labro@iesl.forth.gr

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Abstract. We present photoelectron angular distributions resulting from the two-photon direct double ionization of helium, under XUV radiation, by solving the time-dependent (TD) Schrödinger equation. The helium TD wavefunction is expanded in terms of fully correlated multichannel states normalized with incoming-wave boundary conditions. The present study focuses on fields of pulse durations within the subfemtosecond regime and at photon energy of 45 eV where the direct double ionization channel dominates the sequential channel. In addition, at this photon energy, the ejected electrons, resulting from the direct and the sequential path, acquire non-overlapping kinetic energy spectra. Our study reveals a trend for back-to-back ejection asymmetry independently of the kinetic energies of the electrons, thus implying that angular correlations are taking place at the time of the ionization. In addition, for given kinetic-energy sharing, it appears that this asymmetry is developed within an interatomic time interval of subfemtosecond scale, intimately connected with the electron–electron interaction strength.

\textsuperscript{4} Author to whom any correspondence should be addressed.
Our purpose in this paper is three-fold. Firstly, we present a brief summary of the status of theory in connection with two-photon double ionization (TPDI) and in particular what is referred to as the direct process. Secondly, we provide certain details of the formalism on which our calculations have been based over the last seven years, and which complement our previous publications. Thirdly, we present our new results on photoelectron angular distributions (PADs) pertaining to the direct process.

The basic features of TPDI, and in particular the distinction between direct and sequential TPDI, were pointed out in [1], which provided a quantitative extension of the ideas in a much earlier paper [2]. The crucial result of the work in [2] was the identification of the direct process and the realization that, for a particular range of photon frequencies, it can be distinguished from the sequential process spectroscopically, through the angle integrated photoelectron energy spectrum. This has been followed by papers [1]–[24] with an increasing rate of appearance, reporting more quantitative results, based on a variety of approaches. With the exception of one paper on magnesium [13], the main interest has been focused on helium, as it lends itself to quantitative calculations, as well as cleaner experiments, assuming that the available radiation sources will eventually reach the necessary intensity.

Single-photon double ionization in helium has been studied in great detail over the last 40 years [25–27]. The calculation of the relevant cross-sections, a well-understood problem, requires a satisfactory description of the initial and final states, so that the dipole matrix element between these two states can be obtained, within the desired accuracy. It is well known that for this matrix element to be non-vanishing, correlation must exist either in the initial or the final state [28]. In reality, it is of course present in both, and that is where the challenge in the calculation of double ejection lies. How this aspect is apportioned among the two states, may depend on the method of calculation. Exact calculations, in the true sense of the word, do not exist; although the numerical accuracy of some, for all practical purposes, comes very close. It may be useful to recall at this point that, in a single-photon process, the photon can be absorbed only by one of the electrons. As a result, for double ejection to occur, that electron must interact...
with the other. In the literature on the subject, the whole process is often broken down into two-step, initial state correlation and shake off \[29, 30\]. We do not need to enter into the details of that discussion, but only to note that it underscores the basic fact that only one of the electrons can absorb the photon and that is why correlation is necessary.

The seemingly simple generalization to the TPDI ushers in many layers of difficulty, as well as richness. A satisfactory description of the initial and final states is again necessary but, as in any photoabsorption process beyond single-photon, not sufficient. The physical reason is that the completion of the process involves the successive absorption of more than one photon via intermediate real (if the photon frequency happens to be near resonance with an excited atomic state) or otherwise virtual intermediate states. In either case, it consists of a linear combination of an infinite set of atomic states, where one of them may dominate in the case of near resonance. In the context of lowest non-vanishing order of perturbation theory (LOPT), the calculation of the cross-section must now cope, as in any two-photon process, with the summation over a complete set of intermediate states; an \((N - 1)\)-fold summation for an \(N\)-photon process \[4, 31\]. Obviously, the additional issue of correlation in the intermediate states becomes now as important. How and to what degree this aspect manifests itself in a calculation will surely depend on the basis and the method. Perhaps the relatively easiest part of the calculation is to obtain a satisfactory ground state, which we will assume to be the case.

1.1. Single ionization

The summation over intermediate states represents a demanding task even in the cases of a one-electron atom; or of a single active electron approximation, valid for infrared and optical radiation, in which only one valence electron undergoes the transition. If the Green’s function, or a good approximation thereof, is known, the summation(s) can be performed practically exactly, as is the case for atomic hydrogen \[32\] or an alkali atom within the single-channel quantum defect theory \[33\]. An alternative approach, known as the Dalgarno–Lewis \[34\] or implicit summation method, relies on the solution of a set of differential equations, through which the intermediate state wavefunctions are determined. In principle, its accuracy can be extremely good, limited only by numerical considerations. A third approach, valid for any atom, consists of an explicit summation of a sufficiently large number of terms involving products of matrix elements connecting the initial to the intermediate and the intermediate to the final state, divided by the corresponding energy difference denominators. It is referred to as the truncated summation method and its accuracy is determined by the number of terms included. A result can be considered converged when the inclusion of additional terms in the summation does not change the value of the cross-section beyond the desired accuracy. Clearly, beyond numerical considerations involved in the summation, the quality of the basis employed plays an important role in the success of the calculation. In the very early days of multiphoton processes, in the spirit of seeking qualitative estimates, instead of pursuing convergence through the size of the truncated set, a so-called average energy denominator approach was adopted \[35\], in which all energy differences in each summation were replaced by a single one, in which some energy, roughly equal to that which the intermediate photon reaches, replaced the atomic energies. This average energy difference was pulled out of the summation, in which case the sum of the products of dipole moments, due to the completeness of the set, collapses to a single matrix element of \(r^2\) between the initial and final states; or a matrix element of \(r^{N-1}\), for an \(N\)-photon process. Although, for a two-photon process, for example, there exists an energy for which the
procedure would yield the exact value, there is no way of knowing in advance the value of that energy. As a result, the method was practically abandoned after the late 60s. A fairly thorough review of all those early approaches can be found in [31].

1.2. Double ionization

For a two-electron atom and beyond, any of the above methods would in principle be applicable. The applicability of the truncated summation method is self-evident and requires no further comment here, beyond the obvious reminder that a complete basis for the atom under consideration is understood. This requirement introduces the first major complication in comparison to the one-electron case. Owing, however, to several decades of work on the structure of and photoabsorption in helium, a variety of techniques are available at this point in time, for the construction of such bases. A second complication has to do with the presence of doubly excited states above the first ionization threshold and of course, the double continuum which is the final state for the problem of interest in this paper. A third complication arising in connection with TPDI stems from the fact that the first photon reaches the continuum, which implies that a pole occurs in the summation over intermediate states. This can, however, be handled through existing techniques. All of the above complications may make the task more demanding computationally, but the quantity to be calculated is nevertheless well defined. One of the early calculations of the TPDI cross-section in LOPT was in fact obtained through this approach, using a two-electron basis constructed in terms of B-splines [7]. Very recently, a calculation employing the idea of the average energy denominator, under the name closure, has been published by Kheifets and Ivanov [36]. Among other things, the accuracy of that calculation is subject to the limitation inherent in the method, as discussed above. In a very recent calculation employing B-splines combined with exterior complex scaling, the summation over intermediate states was performed through a technique in the spirit of the implicit summation [37]. To the best of our knowledge, an approach through the Green’s function has not been attempted.

The calculation of the transition amplitude through LOPT presents certain advantages. Unlike the single-photon process, TPDI can in principle occur without correlation anywhere; i.e. even for non-interacting particles. Yet correlation is present in the initial and final, and also in the intermediate states. Again, how it is apportioned among these three stages will depend on the basis. Accepting that the ground state is what it is, a question peculiar to the two-photon case would be: what is the importance of correlation in the intermediate state(s), in addition to that in the final state? It is known that the singly excited states of He exhibit very little correlation. But the linear combination of states that corresponds to the intermediate state, for photon energies above the first ionization threshold, does contain a component of doubly excited states, which can involve significant correlation. It would then be interesting to have the possibility of exploring the degree of importance of that aspect, which would most likely also depend on the photon energy. A truncated summation approach, in terms of a multiconfiguration basis, lends itself to a straightforward exploration of that question, as one can at will manipulate the configuration content at each stage.

The applicability of LOPT, in whatever form, presupposes certain conditions; namely an upper bound on the intensity of the radiation and a lower bound on the pulse duration. The most appropriate calibration of the intensity, for that purpose, is provided by the ponderomotive energy, which contains the necessary information; namely intensity and photon energy. On
the basis of that criterion, LOPT will be valid as long as the ponderomotive energy is significantly smaller than the photon energy; essentially equivalent to the Keldysh parameter being significantly larger than one. Restricting the present discussion to helium, note that for TPDI to be energetically possible, the photon energy has to be larger than 40 eV. Therefore, the intensity would have to be higher than $10^{18}$ W cm$^{-2}$ before the validity of LOPT becomes questionable. As for the pulse duration, it should not be less than, to be on the safe side, 10 cycles of the field. This is necessary for the transition probability per unit time to be meaningful. Given that the sources in that wavelength range available at this time, and expected in the foreseeable future, satisfy both of the above conditions, it is preferably through LOPT that relevant data should be interpreted.

Nevertheless, many calculations over the last few years have been performed through the solution of the time-dependent Schrödinger equation (TDSE) [3, 5, 6, 9, 12, 15–17, 20, 21, 23, 24] and the R-matrix Floquet approach [38]. Actually, a first attempt at a time-dependent (TD) approach, within the so-called essential states approximation, was due to Crance and Aymar quite some time ago [39]. In that paper, they discussed the role of the direct and sequential channels in TPDI. Perhaps the main reason for this preference has been the availability of the necessary formalism and computational tools, developed in connection with the interaction of long wavelength intense lasers with atoms and molecules, under conditions of large ponderomotive energy, which require a non-perturbative approach. In the perturbative regime, where the notion of the cross-section is valid, it can be extracted from the ionization yield obtained from the solution of the TDSE, provided care is taken in accounting for the particular pulse shape and duration employed in the calculation. Due to numerical considerations, however, all TD calculations on this problem have so far been limited to quite short pulse durations, typically 10 cycles or even less. For such short durations, the resulting ionization yield has, not surprisingly, been found to be sensitive to the pulse shape, which has not been the same in all calculations [9, 16]. For this reason alone, cross-sections obtained in calculations for the same intensity may not always be in agreement, even if the rest of the ingredients of the calculation were identical; which is not the case, since the solution of the TDSE, explicitly or implicitly, relies on some basis which differs from calculation to calculation. A further complication arises from the fact that there is no unique and basis-independent way of extracting the direct double ionization signal from the TD wavefunction. Independently of the above uncertainties intruding into TD calculations, a pulse has an inherent bandwidth which may introduce artefacts not present under experimental conditions of much longer duration. For example, a duration of 10 cycles for radiation of photon energy $\sim$45 eV implies a bandwidth of 5 eV. It seems to us that one way to minimize the possible uncertainties or even artefacts in TD calculations is a direct comparison with calculations through LOPT using the same basis.

Although the pulse durations of XUV and soft x-ray sources are for the time being of the order of 30–50 fs or more, calculations for subfemtosecond durations may nevertheless, be useful as a bridge with attosecond physics. Well into the attosecond regime, the time available for the interaction begins approaching the timescale of correlation effects, especially in the intermediate states. In the absence of attosecond sources with sufficient intensity to produce TPDI, such calculations may therefore be of theoretical interest. But it should be kept in mind that their results may not be directly applicable to experimental results under presently available sources.

The present interest in TPDI from the ground state of helium stems from the early realization that it avails itself for the study of the process of direct double ionization not
available under long wavelength (where double ionization relies on what is known as recollision [40, 41]), and it is this process that is sensitive to the various facets of correlation discussed above. Sequential double ionization, proceeding via ionization of He to He$^+$ and the subsequent ionization of He$^+$, will of course always be present to a significant degree. The conditions, namely photon energy and intensity, under which its presence can be minimized have been discussed in some detail elsewhere [18, 19, 22, 42]. Our interest here is focused on the direct process, assuming that, as experimental results already in print seem to suggest [42], there will be ways to single out its signal in ever increasing detail, and in particular PADs, which provide the most sensitive probe of the theoretical model and calculation.

Our first calculation of the total cross-section [7] was based on LOPT and the value obtained was considerably higher than all other values existing in the literature at that time. In the meantime, the recent work by Foumouo et al [20], using a completely different basis in a TD calculation, produced a value quite close to ours. As a cross check, we then performed a TD calculation [24], using our basis, which yielded a result in reasonable agreement with our LOPT value; within numerical uncertainties and the possible limitations due to the small pulse duration. To the best of our knowledge, this still remains the only direct check between an LOPT and a TD result, within the same basis. In the same paper we showed that, if we remove correlation from the final state, we obtain a result in good agreement with [12] who have no correlation in the final state, but surprisingly also with [38] who should have at least some correlation in the final state. In the meantime, other results have appeared with values in between ours (which still remains the highest in print) and the lowest which has been obtained in a close coupling scheme in which closure (i.e. average energy denominator) has been applied [36]. In view of what we know about this method, that result cannot be taken seriously. As for the rest, including ours, time will tell. It should, however, be added that, given the complexity of the problem, it is hardly surprising that disagreements between theoretical results exist, and it will be some time before the issue is resolved.

The new results on PADs reported herein have been obtained through our TD approach, simply because it was more convenient at this time. It is our intention to re-examine them in terms of LOPT in a follow up paper. We would not expect major discrepancies, but it needs to be checked. One possible source of discrepancy could be the bandwidth of our 10 cycles pulse (∼919 as) which spans a ∼5 eV energy band in the continuum, which for two photons amounts to 10 eV, five of which are in the double continuum. As already mentioned above, we begin with certain details of our formal and computational approach in section 2, followed by the results and the relevant discussion in section 3. Finally, we have taken this opportunity to provide some references to the early extensive literature on multiphoton processes.

2. The TD solution framework

2.1. The helium wavefunction in a laser field

The TDSE equation of helium in a TD electromagnetic (EM) field $E = -\partial A(t)/\partial t$ is expressed as,

$$\left[ H + D(p_1, p_2; t) \right] \psi(r_1, r_2; t) = i \frac{\partial}{\partial t} \psi(r_1, r_2; t), \quad (1)$$

where $H$ is the field-free helium Hamiltonian and $D(p_1, p_2; t) = A(t) \cdot p$ ($p = p_1 + p_2$) representing the interaction of the field with the electrons in the velocity gauge and the dipole
approximation. \( \mathbf{p}_1 \) and \( \mathbf{p}_2 \) are the momentum wavevectors of the individual electrons. We assume the expansion of the TD wavefunction on the energy eigenstates of helium as follows:

\[
\psi(r_1, r_2; t) = \sum_{n=1}^{N_B} C_B(t) \Psi_B(r_1, r_2) + \sum_{n=1}^{N_T} C_\Gamma(t) \Psi_\Gamma(r_1, r_2). \tag{2}
\]

The index \( n \) refers to both \( B \) and \( \Gamma \) and should be understood as \( n = B_n \) or \( \Gamma_n \). In the first term, \( \Psi_B \) are the bound states of helium while in the second term \( \Psi_\Gamma \) are the continuum eigenstates. A detailed description of these states will be given in the next two subsections. \( N_B \) and \( N_\Gamma \) are the total numbers of bound and continuum channel states, respectively. Inserting equation (2) into the TDSE we obtain the following coupled system of first-order differential equations for the time-evolution of the coefficients \( C_B(t) \), \( C_\Gamma(t) \):

\[
i \frac{d}{dt} C_n(t) = E_n C_n(t) + \sum_{n'} A(t) \langle \Psi_n | \mathbf{p} | \Psi_{n'} \rangle C_{n'}(t), \quad n = 1, 2, \ldots, N_B + N_\Gamma. \tag{3}
\]

The dipole matrix elements \( \langle \Psi_n | \mathbf{p} | \Psi_{n'} \rangle \) between the two-electron states together with the eigenenergies \( E_n \) are the only dynamical quantities that enter the TDSE. The above system is propagated from an initial time \( t_0 \) up to the final time \( T \) with the EM field assumed vanishing outside this time interval. The initial conditions for the coefficients depend on the state of the atom at the initial time \( t_0 \). After the end of the pulse the TD wavefunction is known, in terms of the coefficients, and all experimental observables are time-independent. Finally, we express the latter observables in terms of the coefficients at the final propagation time \( T \).

In the following, we give the detailed structure of the field-free helium eigenstates used for the expansion of the TD wavefunction and then present the formulas that relate the PADs, shown in the figures, with the TD coefficients.

2.2. Construction of the bound two-electron wavefunctions

The bound two-electron wavefunctions of helium, in a LS representation, are expanded on the following zero-order basis:

\[
\Phi^{(\Lambda)}_{\beta_1}(r_1, r_2) = A_{12} \frac{P_{n_1^{(i)}, l_1^{(i)}}(r_1)}{r_1} \frac{P_{n_2^{(i)}, l_2^{(i)}}(r_2)}{r_2} Y_\Lambda(l_1 l_2; \hat{r}_1 \hat{r}_2), \tag{4}
\]

with \( \beta_i = (n_1^{(i)}, l_1^{(i)}; n_2^{(i)}, l_2^{(i)}) \) and \( i = 1, \ldots, N_\beta \), where \( N_\beta \) is the total number of zero-order two-electron configuration orbitals included in the basis. The operator \( A_{12} \) is the two-body antisymmetrization operator. The functions \( P_{nl}(r) \) are states of the \( \text{He}^+ \) ion, obtained numerically by solving the radial hydrogenic Schrödinger equation (SE) for \( Z = 2 \) for each partial wave, in a grid interval \([0, R]\), subject to boundary conditions \( P_{nl}(0) = P_{nl}(R) = 0 \). The quantity \( Y_\Lambda = Y_{LM_L} Y_{SM_S} \) contains information for the angular and spin quantum numbers of the state \([7]\). We suppress all total and spin angular quantum numbers with the symbol \( \Lambda = (L, S; M_L, M_S) \). Then the two-electron bound wavefunction is given as:

\[
\Psi_B(r_1, r_2) = \sum_{i} C_{\beta_i}^{(B)} \Phi^{(\Lambda)}_{\beta_i}(r_1, r_2), \tag{5}
\]

where \( B = (\Lambda; E) \). The coefficients \( C_{\beta_i}^{(B)} \) represent the configuration interaction (CI) strength between the different configurations \( \beta_i \) that form the total two-electron wavefunction. As
2.3. Construction of the continuum multichannel two-electron wavefunctions

In the present case, we employ the following two-electron basis:

\[
\Phi^{(A)}_{j_1,j_2}(r_1, r_2) = A_{12} \frac{P_{n_1}^{(j_1)}(r_1)}{r_1} \frac{P_{n_2}^{(j_2)}(r_2)}{r_2} Y_{\Lambda}(l_1 l_2; \hat{r}_1 \hat{r}_2),
\]

with \( \gamma_j = (n_1^{(j)}, l_1^{(j)}, l_2^{(j)}), j = 1, \ldots, N_\gamma \) and \( i = 1, 2, \ldots, n_s \). The function \( B_i(r) \) is the \( i \)th member of the B-spline basis defined in an interval \([0, R]\) at grid points given by \( t_i, i = 0, 1, 2, \ldots, n_s + k_s \), with \( n_s \) the number of the B-spline basis and \( k_s \) its order [43]. Then we consider the two-electron multichannel wavefunction expanded as [7]:

\[
\Psi_F(r_1, r_2) = \sum_{j,i}^{N_s, n_s} C_{j,1}^{(A;E)} \Phi^{(A)}_{j_1,j_2} + \sum_k^{N_\beta} C_{\beta_k} \Phi^{(A)}_{\beta_k},
\]

with \( \Phi^{(A)}_{j_1,j_2} \) and \( \Phi^{(A)}_{\beta_k} \) defined by equations (4) and (6), respectively, with \( N_s \) the number of bound zero-order configurations included in the above expansion, not related to \( N_\beta \) of expansion (5).

The two-electron multichannel wavefunction is collectively denoted with the channel index \( \Gamma = (\Lambda; E; \gamma) \). The coefficients \( c_{\gamma,j}^{(A;E)} \) and \( C_{\beta_k} \) being the CI coefficients, are obtained by solving the SE at the desired energy \( E \) (corresponding to the continuum spectrum of the Hamiltonian) as a linear algebraic system of equations. The channel index \( \Gamma \) is associated with the helium continuum eigenstate of total energy \( E \), total angular quantum numbers \( \Lambda \) with one of the electrons left in the ionic state \( \text{He}^+(\epsilon_1, l_1) \) and the other electron possessing angular quantum number \( l_2 \) and asymptotically acquiring momentum equal to \( k_2 = \sqrt{2(E - \epsilon_1)} \). The above wavefunctions, though orthonormal in terms of the angular quantum number \( \Lambda \), since they are calculated in a spherical box, their normalization is box-dependent. As is known, those states can be re-normalized in a number of ways. In the present case we employ a \( K \)-matrix normalization (incoming waves normalization), which has the additional advantage that the dynamical quantities entering the TDSE (with the eigenenergies), namely, the dipole matrix elements, become real quantities.

\[
\lim_{r_2 \to \infty} \Psi_F(r_1, r_2) = \frac{\Phi^{(T)}_{\gamma}(r_1, \hat{r}_2)}{r_2} \left[ \frac{\sin \theta_\gamma(r_2)}{\sqrt{2\pi k_\gamma^{(\gamma)}}} - \sum_{\gamma'} K_{\gamma,\gamma'} \cos \theta_\gamma(r_2) \right],
\]

where \( \Phi^{(T)}_{\gamma}(r_1, \hat{r}_2) \) represents the ionic state plus all the variables of the continuum electron except for the radial variable \( r_2 \). The quantity \( \theta_\gamma(r) \) is defined as:

\[
\theta_\gamma(r) = k^{(\gamma)} r - \frac{\pi}{2} + \frac{Z_{\text{eff}}}{k^{(\gamma)}} \log 2k^{(\gamma)} r + \arg[l_\gamma + 1 - iZ_{\text{eff}}/k^{(\gamma)}].
\]

The effective charge, depending on the target state \( \Phi^{(T)}_{\gamma}(r_1, \hat{r}_2) \) is taken equal to 1 for the single-ejection continuum wavefunction (\( \epsilon_1 < 0 \)) and equal to 2 for double ejection wavefunctions.
3. Differential cross sections for single and double ionization

3.1. Single-ionization

Let us consider the energy-normalized final state with one of the electrons having asymptotic momentum wavevector $k_2$ and the other electron left in the ionic state $\alpha_1 = (n_1 \ell_1)$. We write this state as $\phi^-_{\alpha_1, k_2}(r_1, r_2) = \langle r_1, r_2 | \epsilon_1, l_1; k_2 \rangle$ and given the multichannel state $\Phi_{\Gamma}$, where $\Gamma = (E; \Lambda; \epsilon_1 l_1 l_2)$ and $\epsilon_1 < 0$, we have the following formula that relates the two [44, 45]:

$$\phi^-_{\alpha_1, k_2}(r_1, r_2) = \sum_{\Lambda} \sum_{l_2 m_2} i^{l_2} e^{-i\ell_2(k_2)} Y^*_{l_2 m_2}(\hat{k}_2) C_{\Lambda} \Phi_{\Gamma}(r_1, r_2).$$  \hspace{1cm} (10)

The quantity $C_{\Lambda}$ is equal to $C_{\Lambda} = C_{L M L}(l_1 m_1; l_2 m_2)$ with $C_{L M L}$ the Clebsch–Gordan coefficients. Then the projection operator for single-ionization leaving the ion in the state $P_{\text{SI}}(r)$ and the outgoing electron acquiring asymptotically a wavevector $k_2$ is, $P_{\text{SI}} = \langle \phi^-_{\alpha_1, k_2} | \phi^-_{\alpha_1, k_2} \rangle$. Projection onto the TD wavefunction results in the following expression:

$$\frac{dP_{\text{SI}}}{dk_2}(\hat{k}_2; \alpha_1) = \sum_{\Lambda} \sum_{l_2 m_2} i^{l_2} e^{-i\ell_2(k_2)} C_{\Lambda} Y^*_{l_2 m_2}(\hat{k}_2) \Gamma_{\Lambda}(T) \right|^2,$$  \hspace{1cm} (11)

where $\Gamma_{\Lambda}(T)$ is the TD coefficient present in equation (2). The above equation is evaluated at $k_2 = \sqrt{2(E - \epsilon_1)}$. The unit vector $\hat{k}_2$ defines the direction of the photoelectron, which in the spherical coordinates used here is associated with the angles $(\phi_2, \theta_2)$.

3.2. Double-ionization projection wavefunctions

Let us consider the wavefunction $\phi^-_{\ell_1 \ell_2}(r_1, r_2)$ with the electrons asymptotically having wavevectors $k_1$ and $k_2$. Following Henry and Lipsky [44] we relate the asymptotic wavefunction to the multichannel continuum function $\Phi_{\Gamma}$, with $\Gamma = (\Lambda; E; \epsilon_1 l_1 l_2)$ where $\epsilon_1 = k_1^2/2 > 0$ as:

$$\phi^-_{\ell_1 \ell_2}(r_1, r_2) = \sum_{\Lambda} \sum_{l_1 m_1} \sum_{l_2 m_2} i^{l_1 l_2} e^{-i\ell_2(k_2) - i\ell_1(k_1)} Y^*_{l_1 m_1}(\hat{k}_1) Y^*_{l_2 m_2}(\hat{k}_2) C_{\Lambda} \Psi_{\Gamma}(r_1, r_2).$$  \hspace{1cm} (12)

The projection operator for the doubly-ionized helium is written as $P_{\text{DI}} = \langle \phi^-_{\ell_1 \ell_2} | \phi^-_{\ell_1 \ell_2} \rangle$, and the triply differential cross-section (TDCS) is obtained by projecting onto the TD wavefunction with the result,

$$\frac{dP_{\text{DI}}}{dk_1 dk_2}(\hat{k}_1, \hat{k}_2; \Gamma) = \sum_{\Lambda} \sum_{l_1 m_1; l_2 m_2} i^{l_1 l_2} e^{-i\ell_2(k_2) - i\ell_1(k_1)} C_{\Lambda} Y^*_{l_1 m_1}(\hat{k}_1) Y^*_{l_2 m_2}(\hat{k}_2) \Gamma_{\Lambda}(T) \right|^2.$$  \hspace{1cm} (13)

The above equation is evaluated at $k_2 = \sqrt{2(E - k_1^2/2)}$, $\hat{k}_2$ is defined as in the single-ionization case and the unit vector $\hat{k}_1$ defines the direction of the target photoelectron, which is associated with the angles $(\phi_1, \theta_1)$. 

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4. Results and discussion

In the present work, we have used a pulse of central frequency corresponding to a photon energy \( \omega = 45 \text{ eV} \), peak intensity \( 10^{14} \text{ W cm}^{-2} \) with a sine-squared shape. The maximum number of cycles of the pulse used is 10, corresponding to a pulse duration 919 as. We also assume the field to be linearly polarized considering the polarization axis as the \( z \)-axis. Interaction of the helium atom in the ground state He(1s\(^2\)) with the EM field involves only the singlet \( M_L = 0 \) states. Moreover, if we restrict ourselves to two-photon processes (as the dominant events for the given peak intensity) only states characterized by \( S, P \) and \( D \) total angular symmetry are accessed. Due to the above angular properties of the atom–field system it holds \( \Lambda = (LM_L; SM_S) = (L, 0; 0, 0) \). Therefore any summation over \( \Lambda \) in the related formulas is reduced to a summation over the values \( L = 0, 1, 2 \). In addition, we can safely ignore the spin quantum numbers of the individual electrons since spin–orbit effects are of no importance in the present case. In the present work, we have assumed that the target electron is fixed along the direction of the helium ground state, and \( \theta_1 = 0^\circ \) and \( \phi_1 = 0^\circ \). Then, the angular distribution of the other electron has azimuthal symmetry. In our figures we have set \( \phi_2 = 0^\circ \).

After the propagation of the TDSE from an initial time \( t_0 = 0 \), we obtain the TD coefficients at time \( T \), namely \( C_n(T) \). As initial conditions we set \( C_1(0) = 1 \), where \( n = 1 \) corresponds to the helium ground state, and \( C_n(0) = 0 \) for all other states. Further evolution of the TD wavefunction is under the field-free helium Hamiltonian (time-independent) and its calculation is trivially possible since the TD coefficients satisfy \( C_n(t) = C_n(T) \exp(-iE_n(t - T)) \).

The chosen box-radius was \( R = 40 \text{ au} \) and the number of B-splines \( n_s = 62 \) with their order \( k_s = 9 \). The knot sequence was sine-like [46]. The channels included were of the type ss, pp, sd, dd and pf. As already mentioned, for the construction of the bound and the continuum states we employ somewhat different zero-order bases. More detailed information on the construction of the basis is given in [7, 24].

Turning now to the results we have obtained, in the case of double ejection, the PAD of one of the electrons for fixed ejection of the other electron along the polarization axis (\( \theta_1 = 0^\circ \)) and for various kinetic energy sharing between the electrons is shown in figure 1. Since the photon energy is 45 eV, the total energy of the system after the absorption of the two photons is about \( E = 11 \text{ eV} \). The plotted solid line (a) corresponds to an energy \( \epsilon_1 = 5.98 \text{ eV} \). The dot-dashed line (b) corresponds to \( \epsilon_1 = 2.721 \text{ eV} \) while the dashed line (c) corresponds to an energy sharing with \( \epsilon_1 = 1.36 \text{ eV} \). In reference to this figure, we should point out that for fields of finite duration the total energy at which the system can be found has a peaked distribution around \( E = E_g + 2\omega \) (\( E_g \) being the helium ground state energy), possessing an uncertainty determined mainly by the pulse bandwidth. In the present case, of 10 cycles, the bandwidth is of the order of 5 eV, being of the same order as the kinetic energies of the ejected electrons. This fact, for pulses of attosecond duration and the given photon energy, makes any discussion, in terms of the kinetic energy sharing, of somewhat uncertain value in relation to calculations (not available at this time) with longer pulses or LOPT. The kinetic energy of the second electron is determined now by the relation \( \epsilon_2 = E - \epsilon_1 = E_g + 2\omega - \epsilon_1 \). As is apparent from the figure, in all three cases (a), (b) and (c) a back-to-back ejection asymmetry is always present though its degree varies among them. This suggests that the asymmetry is basically an angular correlation effect [16, 47–49] the degree of which depends on the radial correlation which is related to the kinetic energy sharing between the two electrons. Our present results on the PADS and specifically the tendency towards back-to-back ejection are, to a significant degree, in disagreement with...
Figure 1. PAD for fixed ejection of one (of the two) electron along the polarization axis ($\theta_1 = 0^\circ$) and for different kinetic energy sharing between the electrons. The solid line (a) corresponds to electron kinetic energy $\epsilon_1 = 5.98$ eV. The dot-dashed line (b) corresponds to $\epsilon_1 = 2.721$ eV while the dashed line (c) corresponds to $\epsilon_1 = 2.721$ eV. The total kinetic energy is about $E = 11$ eV.

our early results which indicated more pronounced parallel ejection [8]. At that time, there were no other results to compare to. That calculation was performed in LOPT which we have shown [24] to yield angle integrated cross sections in agreement with the TD approach. As we discovered, in the process of the present calculations, it appears that an erroneous algebraic sign in the contribution of the d-wave had entered those early calculations. In the meantime, results on PADs through alternative approaches [16], as well as general kinematic and symmetry considerations [47, 48], have appeared which also indicate a pronounced tendency for back-to-back ejection. It should perhaps be noted here that kinematic arguments, although extremely valuable in indicating basic features, such as back-to-back emission, need to be supplemented by quantitative calculations, since the degree of back-to-back ejection depends on the photon energy through the value of the coefficients that enter the geometric expression.

In figure 2 again we plot the PAD of one of the electrons for fixed ejection of the other electron along the polarization axis ($\theta_1 = 0^\circ$), after the interaction with a pulse of 10 cycles, but for the various channels included in the calculation of the PAD. Specifically, we show the contribution of channels having individual angular momenta ss, corresponding to $^1S$ states only. As expected the ejection of the second electron is uniformly distributed (solid line) over the entire solid angle ($4\pi$). This is because the spherical harmonics contained in equation (13) are the $Y_{00}$ which have constant value. Next, we plot the PAD for the second electron by including the pp waves, for both $^1S$ and $^1D$ total angular symmetries (dot-dashed line). From this plot, the back-to-back asymmetry is already manifested due to angular correlations between the ss and pp channels. Moreover, the pp waves appears to be a much more dominant process compared to the ss channels for the given laser parameters. This is probably due to the fact that ss waves necessitate correlation both in the initial and the final states, while in the case of the pp waves, correlation only in the final state is necessary, if any. Qualitatively, ss waves can result as
follows: by assuming the $p^2$ CI part of the helium ground state (which contributes considerably less than the $s^2$ configurations) we have: $npn'p + \omega \rightarrow np'cs + \omega \rightarrow e'se + CI \rightarrow e_1se_2s$; while the pp waves can result from the $s^2$ CI part (dominated mainly by the $1s1s$ independent particle configuration) as: $1s1s + \omega \rightarrow 1se + \omega \rightarrow e'pe + CI \rightarrow e_1pe_2p$. Finally by including the sd waves (dashed line, ss + pp + sd) results in a less pronounced back-to-back ejection angle compared to the ss + pp case. This might be due to the fact that CI in the final state for the sd configurations has smaller magnitude than the pp case: $1s1s + \omega \rightarrow 1se + \omega \rightarrow 1se + CI \rightarrow e_1se_2d$. In the former case (pp-wave) the matrix element of relevance is $\langle e_1p | \frac{1}{r_{12}} | e_2p \rangle$, while in the latter case (sd) it is $\langle 1se_1 | \frac{1}{r_{12}} | e_2d \rangle$. In decomposing the above integrals into a combination of one-electron integrals, it should be kept in mind that d-orbitals are subject to a centrifugal potential of higher value than the s- and p-orbitals. Therefore, this centrifugal potential ‘pushes’ the radial wavefunctions of the d-orbitals out of the region where the CI integrands would acquire large values of the quantity $1/r_{12}$. Furthermore, arguing along the above lines, the d-orbital needs to overlap mainly with an orbital (1s) which has all of its probability distributed in a restricted domain of the available radial space, while this is not the case for pp waves where both CI interacting orbitals have their radial probability distributed uniformly in the integral radial domain $(epe', e_1pe_2p)$.

In figure 3 we show a polar plot of the double-ejection PAD for fixed ejection of one of the electrons along the polarization axis (arrow $k_1$, $\theta_1 = 0^\circ$). Arrow $k_2$ points in the direction of the second-ejected electron. Its magnitude gives the relative probability for ejection at different angles. In the figure, we give the results for three different pulse durations $T = 183$ as (dotted line), $T = 367.6$ as (dashed line) and $T = 919$ as (solid line), corresponding to 2, 4 and 10 field cycles, respectively. Since the pulse durations are different, the probabilities for double ejection differ in the three cases. We have scaled the polar plots for the case of two and four cycles so as to coincide with the value of the probability at $\theta_2 = 180^\circ$ for the 10 cycles pulse duration. What is apparent from the figure is that, depending on the pulse duration, the ratio of the ejection probabilities at angles $\theta_2 = 0^\circ$ and $180^\circ$ decreases as the pulse duration increases, making the...
Figure 3. Polar plot of the double-ejection PAD for fixed ejection of one of the electrons along the polarization axis (arrow $k_1$) in the figure for three different pulse durations. Arrow $k_2$ points in the direction of the second-ejected electron. Its magnitude gives the relative probability for ejection at different angles.

back-to-back ejection probability less pronounced. This might be explained by the fact that back-to-back ejection is nothing else than a manifestation of the correlation interaction between the electrons on the way out of the interaction region. On the other hand, the field by itself, acting as a one-body operator, tends to eject the two electrons symmetrically along the polarization axis. It seems therefore that for the given intensity, the reduction of the back-to-back ejection with increasing pulse duration is an indication of the interplay between the static Coulomb interaction between the electrons and the interaction with the field leading to the symmetric ejection of both electrons. Does this mean that, with further increase of the pulse duration, the back-to-back ejection will be reduced even more? Or does it mean that the time window between $\sim 183$ and 900 as represents some characteristic timescale over which electron–electron interaction shapes the final outcome? We would conjecture the second scenario to be the case. If true, which can be decided conclusively only after much more detailed work, then we would have a process whose internal clock is in the attosecond timescale. Clearly, a more thorough analysis of the behaviour of the back-to-back ejection ratio in terms of the field parameters appears desirable, in order to provide deeper insight into the competitive role of the static electron–electron interaction and the TD electron–field interaction.

5. Conclusions

We have provided a sample of PADs expected in two-photon ionization of helium through the direct double ionization channel by radiation of photon energy 45 eV. One general conclusion, which seems to be in overall agreement with other results [16, 17, 47–49], is that the two electrons tend to be emitted in opposite directions. Aside from the approximations involved in any approach to the problem, we have also provided a discussion and specific examples of the possible influence of pulse duration. In exploring that influence, we have found that pulse durations between 100 and 900 as appear to be related to an intra-atomic timescale of
electron–electron correlation. On physical grounds, the existence of such a timescale sounds plausible. The specific time bracket appearing to emerge from our treatment is of course subject to the approximations and limitations of our basis and calculations. Further developments will be needed to clarify this issue, but in any case, it stands to reason that the underlying physics would interface with the attosecond timescale.

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