

Correlation effects in two-photon single and double ionization of helium

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We study the effects of two-photon absorption in the region of the double continuum of helium, with extreme ultraviolet (xuv) laser fields. The nonrelativistic time-dependent Schrödinger equation is solved for the two active electron systems, interacting with strong laser pulses. The dynamic calculations include electronic correlations. The final double continuum states are calculated by treating the electronic term $1/r_{12}$ within the zero- and first-order perturbation theory. We calculate the total single and double ionization probabilities and cross sections for photon energies of 45 and 57 eV, as well as the electron energy distributions in the double continuum. The effects of electronic correlations are discussed in the context of ultrashort pulses.

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I. INTRODUCTION

The development of new extreme ultraviolet (xuv) laser source facilities opens the way to explore light-matter interaction at high frequency, high intensity, and short pulse durations [1,2]. Therefore, it will be soon possible to investigate nonlinear processes in atoms and ions in the xuv regime. It is well known that collective effects often play an important role during laser-matter interactions at short wavelengths. In this case, the single active electron (SAE) approximation is no more valid and the calculations must include channels where both electrons are excited and/or ionized. This increases enormously the computing efforts that are required to calculate the relevant probabilities and cross sections, either by using the lowest-order perturbation theory (LOPT) or by solving the time-dependent Schrödinger equation (TDSE). In this context, the helium atom is the simplest system to test the theory; it is also an excellent candidate for experiments with free electron laser (FEL) sources. Until now, one-photon double ionization ($\gamma, 2e$) of helium has received a lot of attention, from both the experimental and the theoretical sides [3]; theoretical and experimental values now agree fairly well. In the case of nonlinear processes with two active electrons, a great effort has been put on the solution of the TDSE. It is often solved by performing a numerical integration or through spectral type methods (see Ref. [4] for a review and Refs. [5–11] for recent works related to double ionization). A R -matrix Floquet theory has also been successfully used to describe the ($2\gamma, 2e$) process [12]. For two-photon double ionization ($2\gamma, 2e$), the interest has increased in the last few years and several theoretical approaches have been developed. Although the theoretical ($2\gamma, 2e$) cross sections obtained for helium by several groups lie within the same order of magnitude, discrepancies still exist (see Ref. [9] and references therein) and further investigations are needed in this domain. The electron energy distribution spectrum has also received attention from the community. This spectrum is strongly related to the mechanism leading to double ionization: it is generally flat for the direct process

and structured for the sequential one (i.e., when the double ionization results from the ionization of He followed by the ionization of He^+). It is important to recall that, in contrast with one-photon multiple ionization or excitation processes, multiple ionization or excitation resulting from more than one photon absorption does not necessarily require correlations to occur [13]. Therefore, the zero order in perturbation theory in $1/r_{12}$ should give the leading contribution to the ($2\gamma, 2e$) cross sections. In particular, this applies to the isoelectronic series of helium where the zero order in $1/Z$ perturbation theory (PT) becomes exact for large Z (in the nonrelativistic limit) [8]. The aim of this paper is to discuss the correlation effects in direct and sequential two-photon double ionization of helium. A particular attention is paid to the description of the double continuum produced by ultrashort and intense pulses.

We have developed a spectral method of configuration interaction type to solve the time-dependent Schrödinger equation (TDSE) for two active electron systems. We use a technique of discretization with B -spline functions [14] to calculate the whole atomic spectrum. The advantage of B -splines is that they are compact L^2 integrable functions defined in a restricted space usually referred to as a box, in contrast to such L^2 basis set as Gaussian, Sturmian or Slater orbitals that extend to infinity. One of the advantages of solving the problem in a box is that it is easier to control the density of states in the continuum. Nevertheless, by including different pairs of nonlinear parameters, Sturmians have been shown to be efficient to solve the TDSE [8,10]. They also provide very accurate values for high-lying excited states [15]. B -spline expansions are extremely powerful in many situations in atomic and molecular physics (see Ref. [16] for a recent review). For example, they have been recently successfully used to describe the interaction of H_2^+ with strong lasers by using an expansion in terms of prolate spheroidal wave functions confined in an ellipsoidal box [17]. In the present case, products of B -splines are used to represent the radial part of the wave function, which permit to incorporate electron-electron correlations at a high degree of accuracy during the numerical solution of the TDSE. Our approach has been described in previous papers (cited above) and we will not give details here.

Until now the main approximation was to project, at the

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end of the pulse, the solution of the TDSE onto Coulomb states in order to extract the population of the doubly ionized states; it is the zero order approximation of the perturbation theory in $1/r_{12}$. We develop here an approach that partly includes electronic correlations in the double ionization channel. Our method provides quantitative informations for single and double ionization probabilities, as well as electron energy distributions in the double ionization channel.

We present first the theoretical approach. A particular attention will be put on the description of the double continua. Then, we will present results for photon energies of 45 and 57 eV. Note that sequential double ionization, with two-photon absorption, is permitted with photon energies of 57 eV, while this process is only direct in the other case. We will present first cross sections calculated with correlated and uncorrelated double continuum. Besides $(2\gamma, 2e)$ calculations, results will also be given for one-electron ejection with two-photon absorption $(2\gamma, e)$. Then the electron energy distributions will be also analyzed for different representations of the double continuum and with different pulse durations. The calculations are performed at an intensity of 10^{14} W/cm², where three-photon absorption does not play a significant role in the helium case. Atomic units are used throughout the paper unless otherwise mentioned.

II. THEORETICAL APPROACH

A. Atomic structure calculations

Let us first describe the method. The electronic structure of the helium atom satisfies the equation

$$(H - E_n)\Psi_n = 0. \quad (1)$$

H is the nonrelativistic Hamiltonian, which reads

$$H = H_0(1) + H_0(2) + \frac{1}{r_{12}} \\ = -\frac{1}{2}\Delta_1 - \frac{1}{2}\Delta_2 - \frac{Z}{r_1} - \frac{Z}{r_2} + \frac{1}{r_{12}}, \quad (2)$$

where Z is the nuclear charge ($Z=2$). The labels 1 and 2 refer to the electrons 1 and 2, respectively. For a given total angular momentum L and projection M , the solution of Eq. (1) is expanded on a basis of two-electron configurations that are products of one-electron functions as follows:

$$\Psi_n^{L,M}(\mathbf{r}_1, \mathbf{r}_2) = \mathcal{A} \sum_{\alpha \equiv i,j,l_1,l_2} c_{n,\alpha}^{L,M} \frac{B_i(r_1)}{r_1} \frac{B_j(r_2)}{r_2} \mathcal{Y}_{l_1,l_2}^{L,M}(1,2), \quad (3)$$

where \mathcal{A} is the antisymmetrization operator, l_i is the angular momentum of electron i , and $\mathcal{Y}_{l_1,l_2}^{L,M}(1,2)$ is a bipolar spherical harmonic. The B -spline function of order k denoted by $B_i^k(r)$ is a piecewise polynomial of degree $k-1$ [14]. We use N_b B -spline functions, that are distributed along the radial axis, in a radial box defined from $r=0$ to R_{max} . As usual, we choose the B -spline sequence so that $B_1^k(0)=0$ and $B_{N_b}^k(R_{max})=0$ in order to fulfil the boundary conditions. A

direct diagonalization of Eq. (1) gives the eigenenergies and the expansion coefficients $c_{n,\alpha}^{L,M}$ of the bound and continuum states. The cornerstone of our method is that, the system being confined in a box, the whole atomic spectrum is represented by discrete states. Therefore, we do not make any distinction between bound and continuum states and this reduces considerably the numerical difficulties in both perturbative and non-perturbative approaches. Furthermore, it is easy to normalize the continuum states on the energy scale and to extract physical quantities.

B. Time-dependent calculations

Let us 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

$$i\frac{\partial}{\partial t}\Psi_v(\mathbf{r}_1, \mathbf{r}_2, t) = \left[H + \sum_{i=1,2} \mathbf{A}(t) \cdot \mathbf{p}_i \right] \Psi_v(\mathbf{r}_1, \mathbf{r}_2, t). \quad (4)$$

The vector potential, polarized along the z axis, is defined as

$$\mathbf{A}(t) = A_0 \left(\cos\frac{\pi}{T}t \right)^2 \cos(\omega t) \mathbf{e}_z, \quad (5)$$

where T is the total pulse duration, $-T/2 \leq t \leq T/2$, and ω is the photon energy. Similarly, in length gauge, the time-dependent Schrödinger equation reads

$$i\frac{\partial}{\partial t}\Psi_l(\mathbf{r}_1, \mathbf{r}_2, t) = \left[H - \sum_{i=1,2} \mathbf{E}(t) \cdot \mathbf{r}_i \right] \Psi_l(\mathbf{r}_1, \mathbf{r}_2, t), \quad (6)$$

with the electric field defined as

$$\mathbf{E}(t) = -\frac{\partial}{\partial t}\mathbf{A}(t). \quad (7)$$

The time-dependent total wave function is expanded on the basis of the field-free atomic eigenstates, normalized to unity,

$$\Psi_{v,l}(\mathbf{r}_1, \mathbf{r}_2, t) = \sum_{\alpha \equiv n,L}^{N_s, L_{max}} C_{\alpha}^{v,l}(t) \Psi_n^{L,M}(\mathbf{r}_1, \mathbf{r}_2). \quad (8)$$

Equation (4) [or Eq. (6)] is integrated over the total pulse duration T . The initial condition is given by

$$\Psi_l(\mathbf{r}_1, \mathbf{r}_2, t = -T/2) = \Psi_v(\mathbf{r}_1, \mathbf{r}_2, t = -T/2) \\ = \Psi_{in}^{L,M=0}(\mathbf{r}_1, \mathbf{r}_2), \quad (9)$$

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

In order to accelerate the numerical calculations, we eliminate the oscillations of the field-free atomic states by solving the TDSE in the interaction picture. The relation between the total wave function in interaction (I) and

Schrödinger (S) pictures is given, at the end of the pulse, by

$$\Psi_{v,l}^{(I)}(\mathbf{r}_1, \mathbf{r}_2, T/2) = \exp(iHT/2)\Psi_{v,l}^{(S)}(\mathbf{r}_1, \mathbf{r}_2, T/2). \quad (10)$$

C. Description of the final double continuum states

The basis set being not truncated in expansion (3), the electron-electron correlations are included in the solution of the TDSE to a high degree of accuracy. The main approximation comes at $t=T/2$, i.e., when the population of the final double continuum state $\Phi_{klk'l'}^L(\mathbf{r}_1, \mathbf{r}_2)$ is calculated. It is simply given, in the interaction picture, by

$$P^L(klk'l') = |\langle \Phi_{klk'l'}^L(\mathbf{r}_1, \mathbf{r}_2) | \tilde{\Psi}_{v,l}^{(I)}(\mathbf{r}_1, \mathbf{r}_2, t=T/2) \rangle|^2, \quad (11)$$

$\tilde{\Psi}_{v,l}^{(I)}(\mathbf{r}_1, \mathbf{r}_2, t=T/2)$ refers to expansion (8) where the initial state has been excluded [9]. In previous works, we have fully neglected the electron-electron correlations in $\Phi_{klk'l'}^L(\mathbf{r}_1, \mathbf{r}_2)$ and the double continuum was approximated by an antisymmetrized product of hydrogenic functions $\phi_{kl}^m(\mathbf{r}) = R_{kl}(r)Y_l^m(\theta, \varphi)$. This is the zero order of the perturbation theory with $1/Z$ as a parameter, within this approximation the double continuum reads

$$\Phi_{klk'l'}^L(\mathbf{r}_1, \mathbf{r}_2) = \mathcal{N}[R_{kl}(r_1)R_{k'l'}(r_2)\mathcal{Y}_{l,l'}^{L,M}(1,2) + (1 \leftrightarrow 2)]. \quad (12)$$

The normalization coefficient \mathcal{N} insures that

$$\langle \Phi_{klk'l'}^L | \Phi_{klk'l'}^L \rangle = 1 \quad (13)$$

and

$$[H_0(i) - E_{kl}]\phi_{kl}^m(\mathbf{r}_i) = 0, \quad (14)$$

where $i=1$ or 2 and $H_0(i)$ is defined in Eq. (2). For the sake of consistency, Eqs. (1) and (14) are solved using similar B -spline basis set parameters.

We incorporate now first-order correction terms in the expression of the double continuum. We include correlations by diagonalizing the Hamiltonian H in the basis set $\{\Phi_{klk'l'}^L(\mathbf{r}_1, \mathbf{r}_2)\}$ with k, l , and l' fixed. This diagonalization leads to eigenenergies $E_n^{kl'l'}$ and associated eigenvectors $\mathcal{F}_n^{kl'l'}(\mathbf{r}_1, \mathbf{r}_2)$, normalized to unity, which read

$$\mathcal{F}_n^{kl'l'}(\mathbf{r}_1, \mathbf{r}_2) = \sum_{k'=1}^{N_b} C_{n,k'}^{kl'l'} |\Phi_{klk'l'}^L(\mathbf{r}_1, \mathbf{r}_2)\rangle. \quad (15)$$

Note that, if we choose $kl \equiv 1s$, the eigenstates lying above the first ionization threshold represent the $1skl'$ continuum states in the well-known static-exchange approximation. The new representation of the double continuum only involves radial correlations. Screening effects, totally neglected in the zero order perturbation theory, are now partly taken into account in $\mathcal{F}_n^{kl'l'}(\mathbf{r}_1, \mathbf{r}_2)$.

At the end of the pulse, the population of the stationary state $\mathcal{F}_n^{kl'l'}$ with energy $E = E_n^{kl'l'}$ reads

TABLE I. The (l_1, l_2) pairs of angular momenta and total number of combination terms in Eq. (3), for each total angular momentum L . B -spline basis set parameters: $R_{max} = 50$ a.u., $N_b = 50$, $k = 7$, and a linear knot sequence.

Angular momentum L	(l_1, l_2) 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

$$\mathcal{P}_E^L = |\langle \mathcal{F}_n^{kl'l'}(\mathbf{r}_1, \mathbf{r}_2) | \tilde{\Psi}_{v,l}^{(I)}(\mathbf{r}_1, \mathbf{r}_2, t=T/2) \rangle|^2. \quad (16)$$

It is now easy to calculate the population in a double ionization channel (L, l, l') . In the zero order PT [see Eq. (11)], we sum $P^L(klk'l')$ over all k and k' with $E_{kl}, E_{k'l'} > 0$. In the case of the first-order PT calculations [see Eq. (16)], we sum \mathcal{P}_E^L over all n and k values with $E_n^{kl'l'} \geq E_{kl} \geq 0$. In both cases, the total double ionization probability is simply obtained by summing the contributions of the channels $(L=0, l_1, l_2)$ and $(L=2, l_1, l_2)$ with the angular pairs (l_1, l_2) shown in Table I. Note that the eigenenergy $E_n^{kl'l'}$ is the total energy of the two electrons, one of the electrons having the fixed energy E_{kl} , the other electron has the energy $E_{k'l'} = E_n^{kl'l'} - E_{kl}$. The set of energies $E_n^{kl'l'}$ represents a ‘‘smooth’’ double continuum channel and it is easy to normalize the states $\mathcal{F}_n^{kl'l'}(\mathbf{r}_1, \mathbf{r}_2)$ on the energy scale ‘‘ $k'l'$ ’’ by using the density of states [16]

$$\rho(E_{k'l'}) = \rho(E_n^{kl'l'}) = \frac{2}{|E_{n-1}^{kl'l'} - E_{n+1}^{kl'l'}|}, \quad (17)$$

where the labels, $n, n-1$, and $n+1$ refer to the ordering of the eigenenergies $E_n^{kl'l'}$. A similar formula holds for $\rho(E_{kl})$. The density of probability associated to the electron energy distribution reads

$$\frac{d^2 \mathcal{P}_E^L}{dE dE_{kl}} = \rho(E_{k'l'}) \rho(E_{kl}) \mathcal{P}_E^L. \quad (18)$$

We also calculate the one-electron energy spectrum in the double continuum, which is obtained by summing \mathcal{P}_E^L over $E = E_n^{kl'l'}$. In this case, the density of probability reads

$$\frac{d\mathcal{P}_E^L}{dE_{kl}} = \rho(E_{kl}) \sum_{E_n^{kl'l'} \geq E_{kl}} \mathcal{P}_E^L. \quad (19)$$

In order to calculate the total-integrated cross sections, we use the following expressions:

$$\sigma(2\gamma, e) = \left(\frac{\omega}{I}\right)^2 \frac{P_{nk}}{\tau}, \quad (20)$$

TABLE II. The $(2\gamma, 2e)$ cross sections calculated at various photon energies and in interaction (I) and Schrödinger (S) pictures. Cross sections are expressed in $\text{cm}^4 \text{s}$.

ω (eV)	Zero-order PT	First-order PT
45	1.4×10^{-52} (I)	1.7×10^{-52} (I)
	0.9×10^{-52} (S)	0.99×10^{-52} (S)
57	2.4×10^{-52} (I)	2.5×10^{-52} (I)
	2.15×10^{-52} (S)	2.2×10^{-52} (S)

$$\sigma(2\gamma, 2e) = \left(\frac{\omega}{I}\right)^2 \frac{P_{kk}}{\tau}, \quad (21)$$

where I is the intensity in W/cm^2 and the photon energy is expressed in joules. $\tau = \frac{35}{128} T$ (expressed in seconds), the coefficient $\frac{35}{128}$ takes into account the time dependence of the intensity. P_{nk} and P_{kk} are the total probabilities of single and double ionizations, respectively. The single ionization probability P_{nk} is calculated by adding the populations in channels $L=0$ (excluding the initial state) and $L=2$, and subtracting P_{kk} .

III. RESULTS AND DISCUSSION

The B -spline basis set and the configurations used in expansion (3) are given in Table I. The eigenvectors and eigenenergies are calculated by using a standard diagonalization method. In the dynamical part, we include the lowest 1500 eigenstates, for each total angular momentum L . Therefore, there is a total number of 6000 eigenstates in the time-dependent expansion (8). We assume a \cos^2 pulse envelope [see Eq. (5)], the maximum intensity is $I = 10^{14} \text{ W}/\text{cm}^2$ and we choose ten optical cycles for the total pulse duration. We have checked that higher-order processes, such as three-photon ionization, do not play a significant role at this intensity.

Table II shows the cross sections for double ionization of helium through two-photon absorption, for $\omega = 45$ and 57 eV , in the interaction and Schrödinger pictures; we also compare the zero- and first-order PT results. The calculations have been performed in the length gauge, we have checked that the velocity gauge produces similar results. The TDSE equation is solved within the interaction picture by means of an explicit Runge-Kutta method of order 4 [18], we have checked its stability. In principle, i.e., in a complete basis set and with exact stationary states, the results should be inde-

TABLE III. The $(2\gamma, e)$ cross sections calculated at various photon energies and in interaction (I) and Schrödinger (S) pictures. Cross sections are expressed in $\text{cm}^4 \text{s}$.

ω (eV)	Zero-order PT	First-order PT
45	1.6×10^{-51} (I)	1.57×10^{-51} (I)
	1.74×10^{-51} (S)	1.64×10^{-51} (S)
57	8.4×10^{-53} (I)	7.6×10^{-53} (I)
	10.6×10^{-53} (S)	10.1×10^{-53} (S)

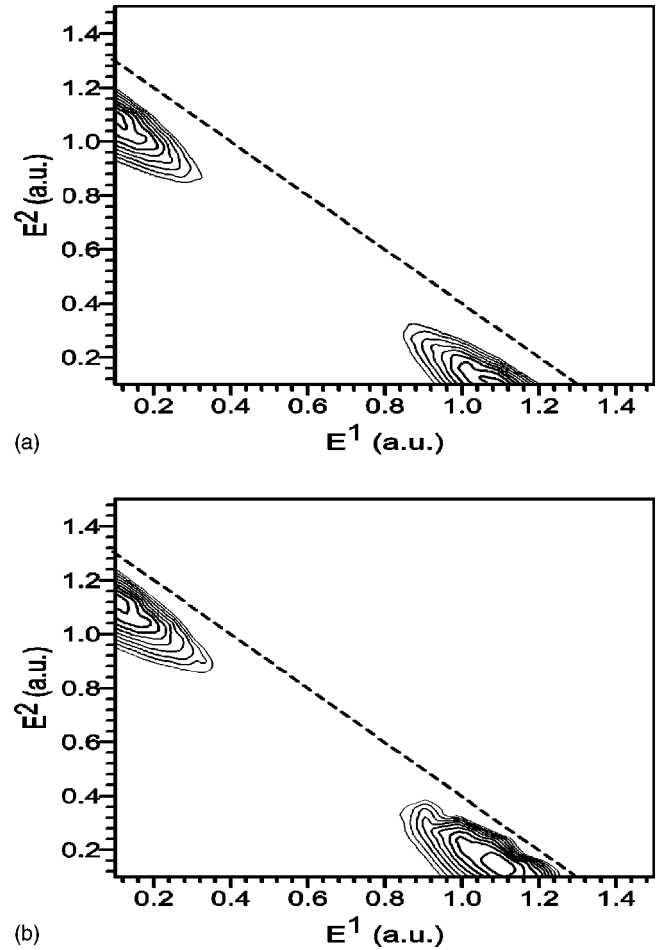


FIG. 1. Comparison of the electron energy distributions between the zero order calculation (a), left, and calculation in first-order of PT (b), right, for $\omega = 57 \text{ eV}$, $I = 10^{14} \text{ W}/\text{cm}^2$, and a pulse duration of 10 optical cycles. The dotted line is the theoretical excess energy $E^1 + E^2 = 1.29 \text{ a.u.}$

pendent of the picture adopted. Nevertheless, due to (i) the truncation in expansion (8) and (ii) the fact that the final states do not have correct standing outgoing conditions, the two pictures lead to different results. Comparison between

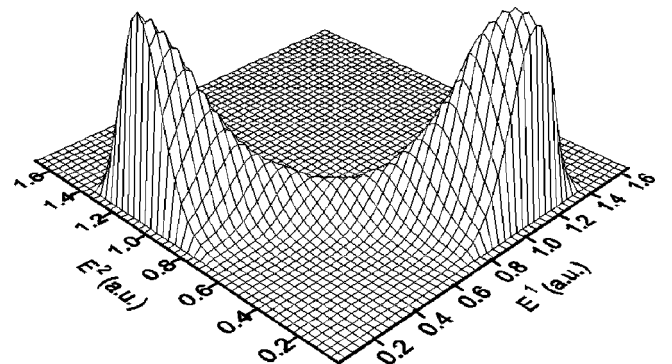


FIG. 2. Electron energy distribution in the dominant channel ($L=2$, $l_1=1$, $l_2=1$), calculated within the first-order PT, for $\omega = 57 \text{ eV}$, $I = 10^{14} \text{ W}/\text{cm}^2$, and a total pulse duration of ten optical cycles.

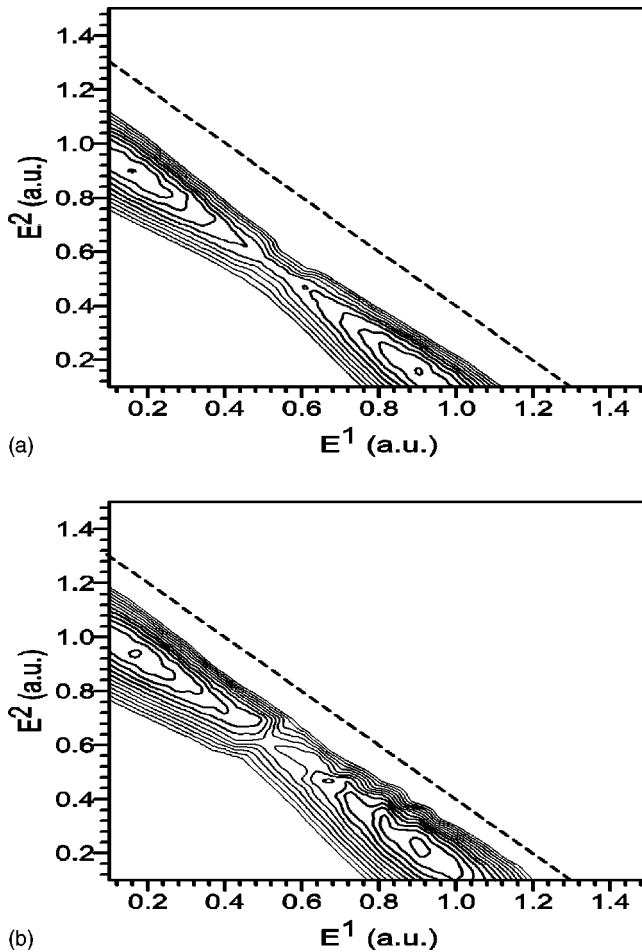


FIG. 3. Same as Fig. 1, with a total pulse duration of six optical cycles.

zero- and first-order PT calculations indicates that for both photon energies electron-electron correlations do not affect significantly the cross sections. Although orders of magnitudes are similar, cross sections calculated in the interaction and Schrödinger pictures differ at 45 eV. At 57 eV, both pictures give similar results. It is also worth noticing that the order of magnitude of the cross sections at 45 and 57 eV are similar.

We have also calculated the $(2\gamma, e)$ ionization cross sections for $\omega=45$ and 57 eV (see Table III). As in the case of $(2\gamma, 2e)$, we consider a total pulse duration of ten optical cycles and a peak intensity of 10^{14} W/cm². Table III shows a good agreement between the zero- and first-order PT results. The interaction and Schrödinger pictures also compare well. In contrast to the $(2\gamma, 2e)$ case, the $(2\gamma, e)$ cross section at 45 eV is much larger than that calculated at 57 eV. Comparing Tables II and III we note that double ionization dominates at 57 eV, while single ionization is predominant at 45 eV.

Besides cross sections, our approach permits to calculate electron energy distributions. In Fig. 1, we have represented the electron energy distribution in the $(L=2, l_1=1, l_2=1)$ channel, which dominates, for a photon energy of 57 eV. The pulse has a total duration of ten optical cycles; its maximum intensity is $I=10^{14}$ W/cm². The distribution is shown in the

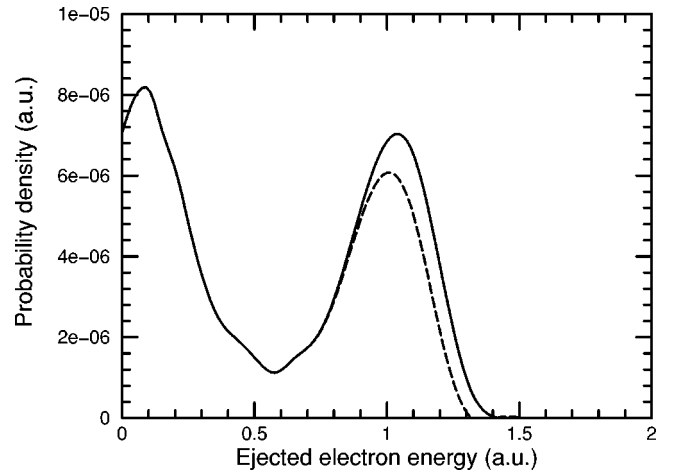


FIG. 4. One-electron energy spectrum in channel $(L=2, l_1=1, l_2=1)$, calculated within the zero order PT (dashed curve) and the first-order PT (solid curve), for $\omega=57$ eV, $I=10^{14}$ W/cm² and a total pulse duration of ten optical cycles.

Schrödinger picture. For the sake of clarity, we have represented only the contour lines which correspond to the higher probabilities. For this photon energy, the theoretical excess energy available in the double continuum is $E^1+E^2 \approx 1.3$ a.u.; it is represented in the figure by a dotted line. We note that the distribution is dominated by two symmetric peaks. Each peak results from a two-step process where the helium atom is ionized through one-photon absorption, then the He⁺ ion is ionized through the absorption of the second photon. During the first step an electron is emitted with the energy $E^1=1.2$ a.u., the second step leads to the ejection of the residual electron with a lower energy, i.e., $E^2=0.1$ a.u. The difference, $E^1-E^2=1.1$ a.u., corresponds to the correlation energy in the ground state of He. Note that the two-step (or sequential) process dominates when the ionization of the He⁺ ion is energetically permitted, i.e., for photon energies larger than 54.42 eV (2 a.u.). This is in agreement with earlier predictions [13]. For lower photon energies (1.45 a.u. $< \omega < 2$ a.u.), the two-photon double ionization process is a direct one and the electron energy distribution is unstructured

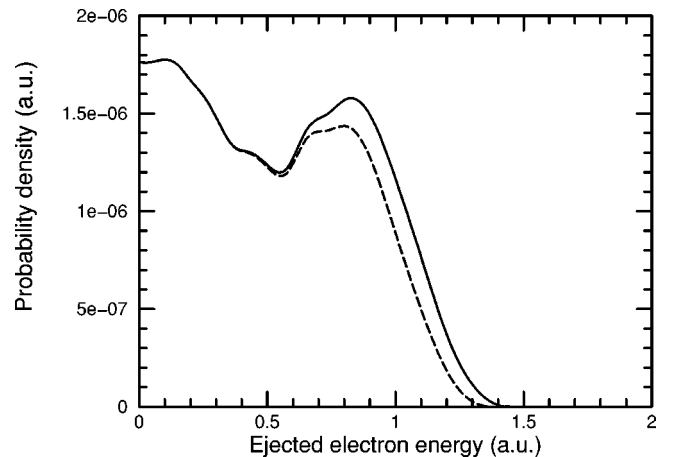


FIG. 5. Same as Fig. 4, with a total pulse duration of six optical cycles.

[8,10]. Zero- and first-order PT calculations are shown in the figure. We remark that the distribution calculated within the first-order PT is closer to the energy conservation line, which indicates that the calculation of the electron energy distribution is improved in the first-order PT. Figure 2 shows the profile corresponding to the case of Fig. 1(b), obtained with the first-order PT. We clearly see the peak at the expected position $E^1 \approx 1.2$ a.u. and $E^2 \approx 0.1$ a.u., and its symmetric. Figure 3 shows a case similar to Fig. 1 but with six optical cycles. The total energy $E^1 + E^2$ is maximum at a value slightly lower than 1.3 a.u. The figure shows that, as in the precedent case, the results are closer to the energy conservation line in the first-order PT. We also note that the maxima get closer to the diagonal, $E^1 = E^2$, for the shortest pulse. Indeed, the pulse duration being of the order of the correlation time of $\text{He}(1s^2)$ (≈ 0.9 a.u.), the core relaxation is not permitted during the double ionization process [8–10]. In the limit of shorter pulses, the double ionization process results from the ionization of two equivalent $1s$ “Hartree” states through two-photon absorption, leading to the production of two electrons having equal energies.

Finally, Figs. 4 and 5 show the one-electron energy spectra obtained within the Schrödinger picture, with the laser parameters of Figs. 1 and 3, respectively. We compare the zero order PT (dotted line) and the first-order PT (full line) results. We clearly see the displacement of the peaks for the shortest pulse duration. We also note that the first-order PT calculations affect the position of the peak located at the highest energy. Indeed, this peak corresponds to the emission of one electron from He (step 1), while the other electron screens the nuclear charge. On the other hand, screening effects are less important in the second step (ionization of the He^+ ion).

IV. CONCLUSION

We have presented a numerical method to treat the problem of multiple ionization of helium by short and intense

laser pulses. Correlations are included in the initial state and during the solution of the TDSE. At the end of the pulse (i.e., at $t = T/2$) the double continuum state is approximated by (1) a simple product of hydrogenic functions and (2) by a function including first-order corrections. Our results show that, as expected, cross sections calculated in zero- and first-order PT do not differ significantly. Nevertheless, the electron energy distribution is influenced by the correlations since the maximum of the distribution is closer to the energy conservation line when the first-order correction is introduced. Indeed, in the latter case, screening effects occurring during the ionization of helium (first step) are partly taken into account. The correlation effects are mainly due to the structure of the initial $\text{He}(1s^2)$ state. For photon energies smaller than 2 a.u., correlations in $\text{He}(1s^2)$ provide part of the required energy for the direct two-photon double ionization process. For higher photon energies, this process leads to two peaks in the one-electron spectrum, separated by the ground-state correlation energy. For ultrashort pulse durations, the electron left in a nonstationary bound state of He^+ has no time to relax and the two peaks move towards each other.

Our objective is to extend our study towards more complex atomic systems, such as beryllium, which requires smaller photon energies to double ionize. The correlation energy in the ground state being much smaller than in He , it can be explored with longer pulse durations. It is of particular interest to study the effects due to the presence of an electronic core.

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