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QUANTUM ELECTRONICS

Photoionization of Helium Atoms through a Superposition of Higher Harmonics

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Abstract. We present a coupled-channel calculation of two-photon single ionization of helium by a superposition of the 7th to the 13th harmonic of a Ti:sapphire laser. Solving the time-dependent two-electron Schrödinger equation with a coherent polychromatic field, the single-ionization probabilities are calculated. Besides Slater-like orbitals we use regular Coulomb wavepackets in our configurational interaction basis to describe the single- and double-electron continuum. Linearly polarized laser pulses are used in the length gauge within the dipole approximation. We applied cosines squared normalized envelope functions. The pulse intensity is varied between 10^9 and 10^{12} W/cm^2, the total duration of each harmonics is between 36–49 femtoseconds. Our results are compared to other *ab initio* calculations, the possible reasons of the discrepancies are discussed.

Keywords: photoionization, higher harmonics, coupled-channel method
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1. Introduction

Multi-photon ionization of atoms induced by short-pulse coherent extreme ultraviolet (XUV) radiations is a challenging problem for a long time now. Recent advances in the high-order harmonic generation (HHG) techniques have led to the development of soft X-ray sources that feature ultrashort pulses with pulse durations of a few hundred attoseconds (as) and may reach intensities ($\sim 10^{14}$ W/cm^2) that are capable of inducing multi-photon processes [1]. Papadogiannis et al. [2] reported the observation of two-photon single ionization of He by a broad band XUV pulse with a spectrum consisting of the 7th harmonic to the 13th harmonic of a Ti:sapphire laser. In our study we analyze this process with our coupled-channel method which will be outlined later.

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Multi-photon, in particular two-photon, ionization of helium by XUV pulses has been studied theoretically by different groups. A considerable numerical effort has been made to solve the two-active electron time-dependent Schrödinger equation (TDSE) with various methods. The configuration interaction B-spline spectral method is one powerful tool [3]. The products of two B-splines represent the radial part of the wavefunction which allows the inclusion of the electron-electron interaction to a high degree of accuracy. Various time-dependent configuration interaction approaches have been used for the interaction with ultra short laser pulses [4, 5], where in [4] time-dependent restricted Hartree–Fock calculations were also presented. A mixed finite-difference basis set technique was employed in [6] to calculate the double-ionization probabilities for short intense pulses. Further details including references and an overview over various methods are given in the review by Lambropoulos et al. [7].

In this paper, we theoretically investigate two-photon ionization of helium by ultrashort femtosecond pulses by solving the TDSE with our coupled channel method which has been originally developed for heavy-ion helium collisions [8–10] and later implemented to describe laser-driven atomic processes and two-photon coherent control [11–13]. To represent bound states and resonances we use Slater-type orbitals. A special feature in our explicitly correlated basis are regular Coulomb wavepackets which we use to discretize the continua. Atomic units are used otherwise mentioned.

2. Theory

The details of our coupled-channel method is introduced in previous works [11, 13] and here we give only a brief summary. To describe the ionization process in the laser pulse we solve the time-dependent Schrödinger equation

$$i\frac{\partial}{\partial t}\Psi(\mathbf{r}_1,\mathbf{r}_2,t) = \left(\hat{H}_{He} + \hat{V}(t)\right)\Psi(\mathbf{r}_1,\mathbf{r}_2,t)\,, \tag{1}$$

where $\hat{H}_{He}$ is the Hamiltonian of the unperturbed helium atom

$$\hat{H}_{He} = \frac{\mathbf{p}_1^2}{2} + \frac{\mathbf{p}_2^2}{2} - \frac{2}{r_1} - \frac{2}{r_2} + \frac{1}{|\mathbf{r_1}-\mathbf{r_2}|}\,, \tag{2}$$

$\hat{V}(t)$ is the interaction operator between the laser pulse and the atomic electrons and will be specified later.

To solve (1) we expand $\Psi(\mathbf{r}_1,\mathbf{r}_2,t)$ in the basis $\{\Phi_i\}$ of eigenfunctions of the time independent Schrödinger equation

$$\hat{H}_{He}\Phi_j = E_j\Phi_j\,, \tag{3}$$

to yield

$$\Psi(\mathbf{r}_1,\mathbf{r}_2,t) = \sum_{j=1}^{N} a_j(t)\Phi_j(\mathbf{r}_1,\mathbf{r}_2)e^{-iE_jt}\,, \tag{4}$$

where the $a_j(t)$ are the time-dependent expansion coefficients and E_j are the eigenvalues in (3). Inserting (4) into the time-dependent Schrödinger equation (1) leads to a system of first-order differential equations for the expansion coefficients

$$\frac{da_k(t)}{dt} = -i \sum_{j=1}^{N} V(t)_{kj} e^{i(E_k - E_j)t} a_j(t) \quad (k = 1, \ldots, N) \tag{5}$$

with the coupling matrix elements

$$V(t)_{kj} = \langle \Phi_k | \hat{V}(t) | \Phi_j \rangle \,. \tag{6}$$

Denoting the ground state with channel $j = 1$ the initial condition before the laser pulse is applied reads

$$\begin{aligned} a_j(t \to -\infty) &= 1 \qquad \text{if} \quad j = 1 \,, \\ a_j(t \to -\infty) &= 0 \qquad \text{if} \quad j \neq 1 \,. \end{aligned} \tag{7}$$

The probabilities for transitions into final helium states j after the pulse are simply given by

$$P_j = |a_j(t \to +\infty)| \,. \tag{8}$$

For the total ionization probability one must sum the P_j which correspond to the discretized channels formed by the wavepackets.

The two-photon single ionization yield is defined via the following expression $W_2 = \hat{\sigma}_2 I_0^2$, where I_0 is the peak intensity of the laser pulse and $\hat{\sigma}$ is the generalized cross section [7]

$$\hat{\sigma}_2 = (\alpha\omega)^2 k P \tag{9}$$

with α being the fine-structure constant, ω the angular frequency of the laser pulse, k the wavenumber of the ionized electron and P the ionization probability.

The eigenfunctions Φ_j in (3) are obtained by diagonalizing the Hamiltonian in a basis of orthogonal symmetrized two-particle functions f_μ so that

$$\Phi_j(\mathbf{r}_1, \mathbf{r}_2) = \sum_\mu b_\mu^{[j]} f_\mu(\mathbf{r}_1, \mathbf{r}_2) \,. \tag{10}$$

In the following we restrict ourselves to singlet helium states only. For the single-particle wave functions we use an angular momentum representation with spherical harmonics $Y_{l,m}$, hydrogen-like radial Slater functions and radial regular Coulomb wavepackets. The Slater function reads

$$S_{n,l,m,\kappa}(\mathbf{r}) = c(n,\kappa) r^{n-1} e^{-\kappa r} Y_{l,m}(\theta, \varphi) \,, \tag{11}$$

where $c(n,\kappa)$ is the normalization constant. A regular Coulomb wavepacket

$$C_{k,l,m,Z}(\mathbf{r}) = q(k, \Delta E) Y_{l,m}(\theta, \varphi) \int_{E_k - \Delta E_k/2}^{E_k + \Delta E_k/2} F_{k,l,Z}(r)\, dk \,, \tag{12}$$

with normalization constant $q(k, \Delta E)$ is constructed from radial Coulomb function of the well-known form

$$F_{k,l,Z}(r) = \sqrt{\frac{2k}{\pi}} e^{\pi\eta/2} \frac{(2\rho)^l}{(2l+1)!} e^{-i\rho} |\Gamma(l+1-i\eta)| \, {}_1F_1(1+l+i\eta, 2l+2, 2i\rho)\,, \quad (13)$$

where $\eta = Z/k,\ \rho = kr$.

The wavepackets cover a small energy interval ΔE_k and thereby form a discrete representation of the continuum which can be incorporated into our finite basis set. The normalized Coulomb wavepackets are calculated up to 315 a.u. radial distance or more to achieve a deviation of less than one percent from unity in their norm. With the help of the Coulomb wavepackets we can make calculations for quiver radii ($r_q = \sqrt{I}/\omega$) of more than 50 a.u. (I stands for the pulse intensity and ω for the photon energy). This would be hardly possible with bound wave functions only.

In our approach two different effective charges Z have been used to take into account the difference between the singly and the doubly ionized electrons. For singly ionized states we have used $Z = 1.0$ and $Z = 2.0$ for the doubly ionized case. A slight deviation from the effective charge gives practically no change in the final spectrum.

Out of the single-particle states (5) we have used 17 s functions (9 Slater functions (sf), 4 wavepackets (wp) with $Z = 1.0$ and 4 wp with $Z = 2.0$), 18 p functions (6 sf, 6 wp with $Z = 1.0$ and 6 wp with $Z = 2.0$) and 12 d functions (4 sf, 4 wp with $Z = 1.0$ and 4 wp $Z = 2.0$) to construct the symmetrized basis functions $f_\mu^{LM}(\mathbf{r}_1, \mathbf{r}_2)$. For the $L = 0$ configurations we have used ss+pp+dd angular correlated wave functions to get a ground state energy of -2.901 a.u. which is reasonably accurate compared to the "exact" value of -2.903 a.u. For the $L = 1, 2$ states we have used only sp or sd configurations. A more detailed analysis of our applied basis can be found in [11, 13].

We restrict ourselves to linearly polarized laser pulses whose coupling to the atomic electrons we describe in the length gauge and in dipole approximation,

$$\hat{V}(t) = -\sum_{i=1,2} \mathbf{E}(t) \cdot \mathbf{r}_i. \quad (14)$$

The laser pulse we use is polarized along the z-axis and has the form of

$$\mathbf{E}(t) = \sum_{N=7,9,11,13} E_N \left(\cos\frac{\pi \cdot t}{\tau_N}\right)^2 [\sin(N\omega_0 t)]\, \mathbf{e}_z\,, \quad (15)$$

where E_Ns are the following relative amplitude ratios of the harmonics $\sqrt{0.4} : 1 : \sqrt{0.4} : \sqrt{0.08}$ according to the experiment of [2]. The total duration at FWHM of each of the harmonics has been assumed to follow the rule of $\tau_N = 130/\sqrt{N}$. The phase shifts between different harmonics are set to zero.

3. Results and Discussion

The coupled-channel equations (5) are solved numerically by using a Runge–Kutta–Fehlberg method of order five, embedding a fourth-order automatic time step regulation algorithm. The results of our theoretical investigation are depicted in Fig. 1 together with the calculations of Papadogiannis et al. [2]. The figure shows the yields of the two-photon single ionization of He by each of the individual harmonics (9th, 11th, 13th and 15th) as a function of its intensity (points) and by the superposition of the harmonics used in the experiment as a function of the total XUV intensity (lines). The original [2] paper consists of many more calculated points for the individual harmonics than listed above. For a better transparency, (or to avoid overlap) we took different pulse intensities in our calculations than [2].

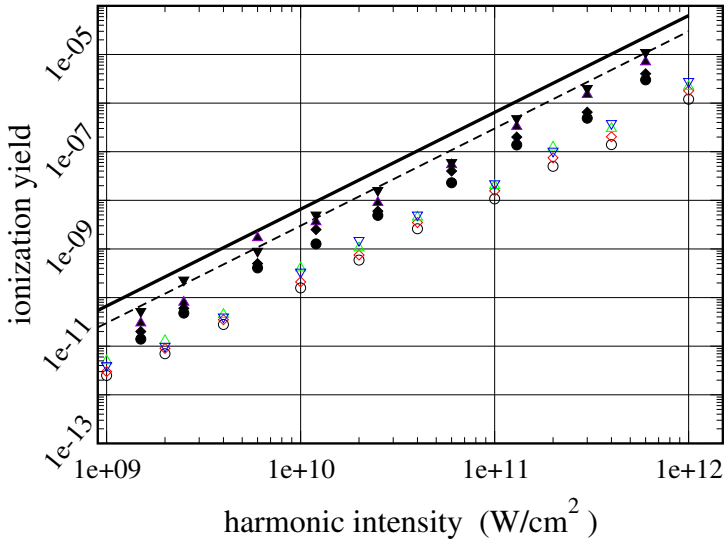

Fig. 1. Calculated yields of He^+ ions as a function of the intensity of the laser filed. The thick solid line represents our results and the thin dashed line is from the work of [2] for the superposition of the 7th, 9th, 11th and 13th harmonic generation. The hollow symbols are data of [2], full symbols represent our calculations. Triangle down: 9th, triangle up: 11th, diamond: 13th and circle: 15th higher harmonic

A careful analysis of [2] data shows that for low pulse intensities 10^9 W/cm^2 the 11th harmonics gives the highest ionization rates, however for moderate intensities (10^{12} W/cm^2) the ionization of the 9th harmonics becomes the highest. It is worth to mention that for individual harmonics the ratio between the largest and the smallest ionization yield is less than a factor of 3. This is a clear fingerprint of a "dense-enough-approximated" electron continuum. Our results compared to

[2] are about a factor of 3 or 5 higher in the whole intensity range for the discrete harmonics. We interpret this phenomena with the intrinsic property of our configuration interaction (CI) basis. It is clear that our full-flagged CI wavefunction describing single-ionized two-electron states contain contributions from bound–bound Slater–Slater orbitals which can not be neglected. Our experience show that there are always some spurious states exist in the single-electron continuum which have larger ionization probabilities than other. These are the ionization states which are overwhelmingly approximated with bound state orbitals rather than Coulomb wavepackets. It is also clear to see that at some intensities the ionization yields of the 9th and 11th harmonics are a factor of 2 or more higher than the corresponding yields of the 13th and 15th. Our explanation is the following, due to linear dependence the size of our basis has a more or less upper limit, and the coverage of the continuum cannot be as dense as for B-splines. This statement is also true for the superposition of the 7th, 9th, 11th and 13th higher harmonics of the Ti:sapphire laser, where our calculation (thick solid line) is about a factor of 4 larger than the data of [2] (thin dashed line).

We should mention that according to the non-perturbative property of our coupled-channel method a large number of different two-photon ionization paths are included for all the four harmonic components. At the level of the coupling matrix (6) the different spectral components of the harmonic fields act independently building up the transition probabilities. However, the time propagation of the coupled-channel equations (5) with N different transition channels describes the complete set of quantum interferences from N different quantum states to the same N quantum states (even for a single harmonics). So quantum interference effects are fully includes in the given basis set. In contrast to perturbative methods the different spectral components can not act independently from each other. With some additional restrictions (e.g. considering that only the first column of the coupling-matrix being non-zero gives back a first order perturbation series) the quantum interference effects can be reduced.

4. Conclusions

We presented results from ab initio coupled-channel calculation for two-photon single ionization of helium by a superposition of the 7th to the 13th harmonic of a Ti:sapphire laser. Linearly polarized laser pulses were used in the length gauge within the dipole approximation. The channel functions were built up from Slater-like orbitals and regular Coulomb wavepackets which helped us to have better physical representation of the single- and double-electron continuum. We applied cosines squared normalized envelope functions. The pulse intensity was varied between 10^9 and 10^{12} W/cm^2, the total duration of each harmonics was between 36–49 femtoseconds. Our results were compared to other ab initio basis calculations, and some discrepancies were found. The possible reasons were discussed. The possible quantum interference effects were analyzed as well.

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