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Enhanced asymmetry in few-cycle attosecond pulse ionization of He in the vicinity of autoionizing resonances

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Received 29 May 2012
Published 13 September 2012
Online at http://www.njp.org/
doi:10.1088/1367-2630/14/9/095010

Abstract. By solving the two-active-electron, time-dependent Schrödinger equation in its full dimensionality, we investigate the carrier-envelope phase (CEP) dependence of single ionization of He to the He\textsuperscript{+}(1s) state triggered by an intense few-cycle attosecond pulse with carrier frequency $\omega$ corresponding to the energy $\hbar \omega = 36$ eV. Effects of electron correlations are probed by comparing projections of the final state of the two-electron wave packet onto field-free highly correlated Jacobi matrix wave functions with projections onto uncorrelated Coulomb wave functions. Significant differences are found in the vicinity of autoionizing resonances. Owing to the broad bandwidths of our 115 and 230 as pulses and their high intensities (1–2 PW cm\textsuperscript{−2}), asymmetries are found in the differential probability for ionization of electrons parallel and antiparallel to the linear polarization axis of the laser pulse. These asymmetries stem from interference of the one- and two-photon ionization amplitudes for producing electrons with the same momentum along the linear polarization axis. Whereas these asymmetries generally decrease with increasing ionized electron

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1367-2630/12/095010+20$33.00 © IOP Publishing Ltd and Deutsche Physikalische Gesellschaft
kinetic energy, we find a large enhancement of the asymmetry in the vicinity of two-electron doubly excited (autoionizing) states on an energy scale comparable to the widths of the autoionizing states. The CEP dependence of the energy-integrated asymmetry agrees very well with the predictions of time-dependent perturbation theory (Pronin et al 2009 Phys. Rev. A 80 063403).

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1. Introduction

The experimental realizations of few-cycle attosecond pulses with stable carrier-envelope phases (CEPs) [1, 2] as well as a proposed method for tuning the attosecond pulse CEP5 represent major milestones toward a main goal of attosecond science: controlling electronic motion on its natural time scale [3–6]. Since the late 1990s there has been an explosion of both experimental and theoretical interest in the CEP effects on electron ionization by sub-10 fs few-cycle laser pulses (see, e.g., [7–20], especially the reviews [9, 14] and references therein). In general, CEP effects on electron angular distributions resulting from ionization by few-cycle laser pulses originate from the interference between amplitudes for ionization by odd and even numbers of photons, i.e. between amplitudes for partial waves of opposite parity [7, 21, 22]. In the case of few-cycle attosecond pulses however, full realization of the goal of controlling electron ionization by means of the pulse CEP requires the experimental development of much more intense attosecond pulses so that nonlinear attosecond processes, as well as attosecond-pump/attosecond-probe processes, can be explored and exploited (for recent experimental advances, see, e.g., [23–27]). Indeed, numerical investigations on asymmetries in electron angular distributions resulting from ionization by isolated linearly polarized few-cycle attosecond pulses showed that such asymmetries only occur for attosecond pulse intensities of

5 On p 445 of [1], the authors state: ‘The attosecond CEP could be finely tuned, for example, by using aluminum foils with variable thickness: At 36 eV, a π CEP variation is induced by the addition of ≈ 80 nm of aluminum (although in this case also the pulse duration is affected).’
the order of 1 PW cm\(^{-2}\) or higher [17, 19, 20]. Moreover, analysis of the intensity dependence of the difference between electrons ionized in opposite directions along the polarization axis of a few-cycle attosecond pulse showed that this difference scales as the 3/2 power of the peak laser pulse intensity, implying that the asymmetry stems from interference of ionization amplitudes for one- and two-photon processes [19]. This implication was confirmed by the perfect agreement between the numerical predictions of [19] and the \textit{ab initio} parameterization of the ionized electron angular distributions provided by a perturbation theory analysis of transform-limited, few-cycle attosecond pulse photoionization in which only terms up to second order in the pulse electric field were included [28]. (Similarly, good agreement between numerical results for asymmetric electron angular distributions produced by chirped few-cycle attosecond pulses and the results of an \textit{ab initio} parameterization of the ionized electron angular distributions provided by a perturbation theory analysis for the case of chirped pulses has also recently been demonstrated [29].) Note that in all these experimental and theoretical investigations of CEP effects on electrons ionized by few-cycle laser pulses, so far there has been no evidence for explicit two-electron effects.

Theoretical interest in the role of autoionizing states in intense laser–atom processes has grown in recent years [30–45], with most of these investigations focused on the time evolution of an autoionization process. Several of the works investigate the role of autoionization in high-order harmonic generation [36, 38, 40, 44]. Only a few of the theoretical investigations deal with the role of autoionization in laser ionization of the He atom [31, 33, 35, 37, 41, 45], but none of these concern the CEP effects on ionization by few-cycle, intense attosecond pulses.

In this paper, we investigate the CEP effects on single ionization of the He atom by a few-cycle, intense attosecond pulse, focusing on ionized electron spectra in the vicinity of the lowest autoionizing states below the He\(^+\)(\(n = 2\)) threshold. The quantum dynamics of highly correlated two-active-electron systems driven by a few-cycle attosecond extreme ultraviolet laser pulse is a fundamental problem in attosecond physics that poses a significant numerical challenge for theory. Helium is the most fundamental system for research on such two-electron correlations and its lowest-energy autoionizing states have quite short lifetimes (for example, 2s2p(\(^1\)P\(^o\)) has a lifetime of \(\tau = 17.7\) fs). For single-cycle attosecond pulses such as those that have been obtained experimentally [1, 2], the pulse bandwidth has the same order of magnitude as the energy corresponding to the carrier frequency. Consequently, for intense pulses there exist ionized electron energies at which one- and two-photon amplitudes interfere. Owing to the different parities of the ionized electron states produced by such interfering amplitudes, asymmetries in the ionized electron yields for ionization parallel and antiparallel to the direction of linear polarization of the laser pulse are found. Our focus is on ionized electron energies in the vicinity of autoionizing resonances, where we find greatly enhanced asymmetries for electrons ionized in opposite directions. We calculate the two-electron wave packet amplitude resulting from interaction of the He atom with a one- or two-cycle attosecond pulse having a carrier frequency of 36 eV, which is comparable to the ones realized experimentally in [1] but with a much larger peak intensity of either 1 or 2 PW cm\(^{-2}\). Our results are obtained by solving the two-electron, time-dependent Schrödinger equation (TDSE) in its full dimensionality for a linearly polarized pulse and we examine the dependence of our results on the CEP. At the end of the laser pulse, the two-electron wave packet is projected onto field-free Jacobi matrix states that are eigenstates of the correlated, field-free Hamiltonian for the two-electron He atom.
In section 2, we provide the essential details of our theoretical approach. In section 3, we present our numerical results and analysis. Finally, in section 4, we briefly summarize our results and draw some conclusions. Note that we employ atomic units \((e = \hbar = m = 1, c = 1/\alpha\), where \(\alpha\) is the fine structure constant) throughout this paper, unless otherwise stated.

2. Theoretical approach

For the few-cycle attosecond pulses considered in this paper, the ionization cross section is not meaningful owing to the broad bandwidth of the pulse. The relevant observable for this paper is instead the probability of producing an electron in the continuum with the residual ion remaining in its ground state, i.e. \(\text{He}^+(1s)\). This probability is obtained by projecting the two-electron wave packet (after the interaction with the pulse) onto the field-free correlated Jacobi matrix (CJM) wavefunction (see, e.g., \([46–53]\)). The CJM wavefunction describes with high accuracy the correlated single continuum system, \(e^- + \text{He}^+(n \geq 1)\), including all processes involving both single ionization without excitation and single ionization with excitation. The role of electron correlations is determined by comparing projections of the two-electron wave packet onto the correlated CJM wavefunctions with the corresponding projections onto uncorrelated Sturmian Coulomb (USC) wave functions \([52]\).

In this section, we first describe briefly our procedure for solving the TDSE. We then describe the CJM approach. Finally, we specify how to extract the differential probability densities and related observable quantities.

2.1. Two-active-electron time-dependent wave packet

In order to investigate the dynamics of a two-electron quantum system (in which the electrons move in a central potential with nuclear charge \(Z\)) that interacts with a linearly polarized, intense laser pulse (within the usual electric dipole approximation), we employ two different accurate methods to solve the TDSE in its fully dimensionality. For both methods, the fully six-dimensional (6D) TDSE has the following form:

\[
\begin{align*}
\frac{\partial}{\partial t} \tilde{\Phi}(r_1, r_2|t) &= \left[ -\frac{1}{2} \left( \frac{\Delta_{r_1} + \Delta_{r_2}}{r_1} - \frac{2}{r_2} + \frac{1}{|r_1 - r_2|} + E(t) \cdot (r_1 + r_2) \right) \right] \tilde{\Phi}(r_1, r_2|t),
\end{align*}
\]

where \(r_1\) and \(r_2\) are the position vectors of each electron with respect to the nucleus, which is located at the origin. The last term in the above equation gives the attosecond pulse interaction with the electrons in the dipole length form. A more tractable form of the 6D wavefunction \(\tilde{\Phi}(r_1, r_2|t)\) is obtained by adopting the close-coupling expansion \([54–56]\) in bipolar spherical harmonics \(\Lambda_{l_1, l_2}^{L M}(\hat{r}_1, \hat{r}_2)\):

\[
\tilde{\Phi}(r_1, r_2|t) = \sum_{LM} \sum_{l_1 l_2} \tilde{\Psi}_{l_1 l_2}^{L M}(r_1, r_2|t) \frac{\Lambda_{l_1, l_2}^{L M}(\hat{r}_1, \hat{r}_2)}{r_1 r_2},
\]

where \(LM\) defines the orbital angular momentum state of the system and the bipolar harmonic, given by

\[
\Lambda_{l_1, l_2}^{L M}(\hat{r}_1, \hat{r}_2) = \sum_{m_1, m_2} \langle l_1 m_1 l_2 m_2 | LM \rangle Y_{l_1, m_1}(\hat{r}_1) Y_{l_2, m_2}(\hat{r}_2),
\]
couples the two individual electron orbital angular momenta $l_1$ and $l_2$ in the $LS$-coupling scheme, where $Y_{l,m}(\hat{r})$ denotes a spherical harmonic and $\langle l_1 m_1 l_2 m_2 \rangle LM$ is a Clebsch–Gordan coefficient. In order to preserve parity, which is a good quantum number, the orbital angular momenta of the electrons must satisfy $(-1)^{l} = (-1)^{l_1 + l_2}$ for interaction of the ground state with linearly polarized photons. Furthermore, both the Coulomb potential $1/|\mathbf{r}_1 - \mathbf{r}_2|$ and the electric dipole interaction terms can be expanded in terms of spherical harmonics. Substituting these expansions into the Schrödinger equation (1) and integrating over the angles $\hat{r}_1$ and $\hat{r}_2$ yields a set of coupled partial differential equations with only two radial variables $r_1$ and $r_2$:

$$i \frac{\partial}{\partial t} \tilde{\Psi}(r_1, r_2|t) = \left[ \hat{T}_1 + \hat{T}_2 + \hat{V}_C^1 + \hat{V}_C^2 \right] \tilde{\Psi}(r_1, r_2|t) + \sum_k \hat{V}_{j,k}^I(r_1, r_2|t) \tilde{\Psi}_k(r_1, r_2|t),$$

(4) where the partial wave index $j$ runs from 1 to the total number $N$ of specific orbital angular momentum combinations $(L, M, l_1, l_2)$ employed in the expansion (2). In equation (4), the diagonal operators $\hat{T}_1$, $\hat{T}_2$ and $\hat{V}_C^1$, $\hat{V}_C^2$ give, respectively, the kinetic energy of each electron and the Coulomb interaction of each electron with the nucleus, while the off-diagonal potential term $\hat{V}_{j,k}^I(r_1, r_2|t)$ includes the Coulomb repulsion between the two electrons and their interactions with the external field. Since we assume that the attosecond pulse is linearly polarized along the $z$-axis and that the electric dipole approximation is valid, the magnetic quantum number takes the value $M = 0$ if the He atom is initially in its ground state.

One method we have used to numerically solve the above coupled partial differential equations (4) is the recently developed TDSE parallel solver (RSP-FE-DVR), designed to run on high-performance supercomputers [57, 58]. This method combines the real-space product (RSP) algorithm with the fine-gridding scheme of the finite-element discrete-variable representation (FE-DVR). To study the ionization of He by an attosecond pulse, we first propagated a trial wave packet with eight partial waves ($l_1 = l_2 = 0 - 7$ and $L = 0$) in imaginary time to obtain the He ground state. In subsequent calculations for the interaction dynamics of the He ground state with an attosecond pulse, we employed a total of 43 partial waves in the expansion, with the total angular momentum $L$ taking values from 0 to 3, and including all possible combinations of $l_1 = l_2 = 0 - 7$ for each value of $L$. We employed a two-dimensional grid of 120 Bohr radii, spanned by 60 finite elements. An eight-point basis is used within each equal-sized (2.0 Bohr) finite element. In addition, we enforced an absorption edge for large $r_1$ and $r_2$ boundaries to avoid wave reflections from the outer boundaries.

The second method we have used to numerically solve the coupled partial differential equations (4) has recently been described in [59]. The radial wave function, $\tilde{\Psi}_{(LM)}(r_1, r_2|t)$ (cf equation (2)), is discretized using the normalized Gauss–Lobatto FE-DVR basis functions $[60, 61]$, whose advantages, detailed in [59], have been previously demonstrated by many other authors $[62–64]$. The matrix elements of the electron–electron Coulomb repulsion term in the Hamiltonian need to be handled especially carefully; our strategy for this follows the treatment reviewed by McCurdy et al $[65]$. The solution of the TDSE (4) is obtained using an effective iterative Arnoldi–Lanczos method $[66]$, whose accuracy and stability have been verified in a number of recent works $[67–70]$. We note that the above two different methods for solving the two-electron TDSE give essentially the same converged results for the two-electron wave packet, as determined by their projections onto both correlated and uncorrelated field-free two-electron states, which are described in the next subsection.

2.2. Helium field-free correlated Jacobi-matrix wave functions

In this work, the field-free, singly ionized continuum states of helium are described by a multichannel scattering wavefunction which can be generated accurately using the so-called Jacobi- or $J$-matrix method [46–53]. This method, which is a type of Galerkin spectral method, bears a close resemblance to the $R$-matrix theory (cf [49]). As in that theory, the configuration space is divided into two regions. In the inner region, the space is spanned by the same finite Coulomb–Sturmian basis used to generate the eigenstates of the Hamiltonian. In the outer region, it is assumed that the outgoing electron moves in a screened Coulomb potential. In order to reproduce correctly the asymptotic behavior of the outgoing-electron wave function in each channel, it is expanded in an infinite basis of Coulomb–Sturmian functions. We use the Greek letters $\gamma, \lambda, \mu$ for the radial and angular quantum numbers of the bound electron, and the Roman letters $n, l$ and $m$ for the Sturmian index and the angular momentum quantum numbers of the ejected electron. For a given channel $\Gamma$, a given outgoing-electron energy $E = k_\gamma^2/2$, and total energy $E_\gamma$, the CJM wave function may be written as

$$\Theta_\Gamma(\mathbf{r}_1, \mathbf{r}_2, E_\gamma, \hat{k}_\gamma) = \sum_j b_j^\Gamma(E_\gamma, \hat{k}_\gamma) \Psi_j^{L,M}(\mathbf{r}_1, \mathbf{r}_2) + \sum_{\Gamma'} \sum_{n=0}^\infty f_{n}^{\Gamma\Gamma'}(E_\gamma, \hat{k}_\gamma) \Phi_n^{\Gamma'}(\mathbf{r}_1, \mathbf{r}_2),$$

where the channel $\Gamma \equiv (\nu, \lambda, \mu; L, M)$ designates the target radial and orbital angular momentum quantum numbers, the orbital angular momentum of the ejected electron and the total orbital angular momentum and its projection respectively. In equation (5), the two-electron function $\Psi_j^{L,M}(\mathbf{r}_1, \mathbf{r}_2)$ in the inner region is expanded in Sturmian functions, as follows:

$$\Psi_j^{L,M}(\mathbf{r}_1, \mathbf{r}_2) = \sum_{l_1,l_2} \sum_{n_1,n_2} \alpha_{l_1,l_2}^{n_1,n_2} \psi_{l_1,l_2,L,M}^{n_1,n_2,j} A \left( \frac{S_{n_1,l_1}^k (\mathbf{r}_1)}{r_1} \Lambda_{l_1,l_2}^{L,M} (\hat{r}_1, \hat{r}_2) \frac{S_{n_2,l_2}^k (\mathbf{r}_2)}{r_2} \right),$$

where the expansion coefficients $\psi_{l_1,l_2,L,M}^{n_1,n_2,j}$ are obtained by requiring that the wave function $\Psi_j^{L,M}(\mathbf{r}_1, \mathbf{r}_2)$ solves the time-independent Schrödinger equation. In fact, by performing a single diagonalization of the atomic Hamiltonian for each value of the total angular momentum $L$, we obtain discrete eigenenergies corresponding to bound states and to pseudostates representing the continuum. The operator $A = (1 + \epsilon P_{1,2})/\sqrt{2}$, where $P_{1,2}$ is the electron interchange operator, projects onto either singlet ($\epsilon = +1$) or triplet ($\epsilon = -1$) states in order to ensure the symmetry or antisymmetry of the spatial wave function, as required by the Pauli principle. The coefficients $\alpha_{l_1,l_2}^{n_1,n_2} = 1 + (1/\sqrt{2} - 1)\delta_{l_1,l_2}^{n_1,n_2}$ control the redundancies (or normalization) that may occur in the basis under interchange of the electrons. The radial hydrogen-like functions $S_{n,j}^k(r)$ are Coulomb–Sturmian functions (defined in detail in [52]), which form a complete discrete basis of square-integrable functions with $\kappa$ as the so-called dilation parameter. The angular part of the expansion (6) is expressed in terms of bipolar harmonics (3).

The right-hand side of (5) contains two terms: the first term represents the scattering wavefunction in the inner region, while the second term describes its asymptotic behavior by a double expansion over all channels $\Gamma'$ and over $n'$, the radial index of the Coulomb–Sturmian functions describing the ejected electron. The two-electron wavefunction in the outer region, $\Phi_n^{\Gamma'}(\mathbf{r}_1, \mathbf{r}_2)$, is given by

$$\Phi_n^{\Gamma'}(\mathbf{r}_1, \mathbf{r}_2) = A \left( \frac{\chi_{\nu',k'}^k(\mathbf{r}_1)}{r_1} \Lambda_{\nu',l',j}^{L,M} (\hat{r}_1, \hat{r}_2) \frac{S_{n',l'}^k (\mathbf{r}_2)}{r_2} \right),$$

where $\chi_{\mu \lambda}^\nu(r_1)$ is the inner electron wavefunction associated with either a bound state (for negative energies) or a pseudostate in the continuum of the residual ion (for positive energies). In order to facilitate the numerical computation of matrix elements, $\chi_{\mu \lambda}^\nu(r_1)$ is obtained by diagonalizing the Hamiltonian for He$^+$ in our finite Coulomb–Sturmian basis. The coefficients $b_n^\nu(E_t, \hat{k}_e)$ and $f_n^\nu(E_t, \hat{k}_e)$ in equation (5) are obtained by requiring that $\Theta_\Gamma(r_1, r_2, E_t, \hat{k}_e)$ solves the Schrödinger equation, giving an algebraic system of equations that these coefficients must satisfy. The outer region expansion coefficients are written as follows:

$$f_n^\nu(E_t, \hat{k}_e) = f_n^\nu(E_t) \sum_{\mu' m'} \langle \lambda', \mu' m' | L M \rangle Y_{l' m'}(\hat{k}_e),$$

(8)

where the angular dependence is described by the Clebsch–Gordan coefficient and the spherical harmonic $Y_{l' m'}(\hat{k}_e)$, with $\hat{k}_e \equiv (\theta, \phi)$. In order to calculate the angular distribution of the ionized electron, we must sum coherently over the angular momentum quantum numbers of the continuum electron and its coupling to the ion core. For this reason, we define

$$\Theta_\Gamma(r_1, r_2, E_t, \hat{k}_e) = \sum_{LM} \sum_l \Theta_\Gamma(r_1, r_2, E_t, \hat{k}_e),$$

(9)

where $\hat{\Gamma} \equiv (\nu \lambda)$ denotes only the quantum numbers of the bound ion state. The summations over $LM$ and $l$ in equation (9) and over $\Gamma'$ in equation (5) lead to interference between odd- and even-photon transition amplitudes (and hence to asymmetries in the ionized electron angular distribution in the case of a few-cycle pulse). The term $f_n^\nu(E_t)$ in (8) describes the asymptotic boundary condition

$$f_n^\nu(E_t) = \gamma_n^\nu(E_t - \epsilon) \delta_{\Gamma \Gamma'} - \Xi_n^\nu(E_t - \epsilon) T_{\Gamma \Gamma'},$$

(10)

where $l'$ belongs to the channel $\Gamma'$, $\epsilon$ is the energy of the electron that remains bound and $\gamma_n^\nu$ is the expansion coefficient of the regular Coulomb wavefunction $\psi_k^\nu(r_2)$ (satisfying incoming-wave boundary conditions) in Sturmian functions. The Coulomb function $\psi_k^\nu(r_2)$ for the ionized electron together with $\chi_{\mu \lambda}^\nu(r_1)Y_{\lambda', \mu'}(\hat{r}_1)$ for the bound electron, appropriately symmetrized, describe the singly ionized continuum of helium by including only screening effects (i.e. the first order of the multipolar expansion of $1/r_{12}$ [44]). The corresponding wavefunction, which includes only the first term on the right-hand side of (10), is thus an uncorrelated Sturmian–Coulomb (USC) wave function. The expansion coefficient $\Xi_n^\nu$ in the second term on the right-hand side of (10) represents either an outgoing Coulomb wave for open channels (with $k^2_e = 2(E_t - \epsilon) > 0$) or a dying exponential for closed channels (with $k^2_e = 2(E_t - \epsilon) < 0$). We emphasize that closed channels do contribute to the transition matrix $T_{\Gamma \Gamma'}$. We also mention that the double-continuum channels are treated in an approximate way. Asymptotically, the outer electron is described by a Coulomb wave and the inner one by a pseudostate. In the inner region however, the double continuum is described correctly. The total energy $E_t$ of the system for a $q$-photon-induced, (possibly) core-excited, single-ionization process is related to the ionized electron energy $E$ and to the carrier frequency $\omega$ of the laser pulse by energy conservation, $E_t(\text{au}) = -2.9037 + q \omega = E + \epsilon$.

2.3. Differential ionization probabilities

To obtain the probabilities for single ionization to a particular state $\tilde{\Gamma}$ of the ion, we must project the continuum part, $\Phi_{\Gamma}(t, \hat{k}_e)$, of the two-electron wave packet $\Phi(t)$ in equation (2) (after the interaction
with the laser pulse) onto the field-free CJM wave functions $\Theta_{\tilde{\Gamma}}$ defined in equations (9) and (5). Note that $\Phi_C \equiv \Phi - \Phi_{BS}$ is the continuum part (including both single and double continua) and $\Phi_{BS}$ is the bound part of the total wave packet $\Phi$. For a linearly polarized, few-cycle laser pulse with a CEP $\varphi$, the axial symmetry of the problem allows us to express the differential probability density $P_{\tilde{\Gamma}}(E, \theta, \varphi)$ for ionization to a particular ion state $\tilde{\Gamma}$ by

$$P_{\tilde{\Gamma}}(E, \theta, \varphi) = |\langle \Phi_C(\varphi)|\Theta_{\tilde{\Gamma}}(E, \hat{k}_z) \rangle|^2.$$  

(11)

For the electrons ionized along the $z$-axis ($\theta_+ = 0$ or $\theta_- = \pi$), upon integrating the differential probability density over the energy of the ionized electron, one obtains information about the CEP dependence of the energy-integrated differential probability density

$$P_{\tilde{\Gamma}}^\pm(\varphi) = \int_0^\infty P_{\tilde{\Gamma}}(E, \theta_\pm, \varphi) dE.$$  

(12)

In order to study the asymmetry in ionization of electrons along the $z$-axis let us define, as in [19, 28], the energy-dependent asymmetry $\Delta P_{\tilde{\Gamma}}(E, \varphi)$,

$$\Delta P_{\tilde{\Gamma}}(E, \varphi) \equiv P_{\tilde{\Gamma}}(E, \theta_+, \varphi) - P_{\tilde{\Gamma}}(E, \theta_-, \varphi),$$  

(13)

and the energy-integrated asymmetry $\Delta P_{\tilde{\Gamma}}(\varphi)$,

$$\Delta P_{\tilde{\Gamma}}(\varphi) \equiv P_{\tilde{\Gamma}}^+(\varphi) - P_{\tilde{\Gamma}}^-(\varphi).$$  

(14)

In the remainder of this paper, we drop the channel index $\tilde{\Gamma}$ in the asymmetry parameters in equations (13) and (14) since $\tilde{\Gamma} = 1s$ is the only ion state for which we have calculated these parameters.

3. Numerical results

Our numerical results on asymmetries in the single ionization of He by an intense, few-cycle attosecond pulse are presented and discussed here. In the first subsection, we specify the parameters of the few-cycle attosecond pulses we employ in this work. We also discuss our general considerations for choosing the parameters of these pulses. In the second subsection we compare our correlated and uncorrelated results for ionization of electrons parallel and antiparallel to the $z$-axis (the direction of linear polarization of the pulse) over a large energy region from the He$^+(1s)$ threshold to 70 eV above, which includes the region of doubly excited, autoionizing states. In the third subsection, we focus on the energy region of the lowest-energy doubly excited autoionizing states, in which we find significant enhancements in the asymmetry for single ionization, parallel and antiparallel to the $z$-axis. In the fourth subsection, we provide our results on the asymmetry in the energy-integrated single-ionization probabilities parallel and antiparallel to the $z$-axis.

3.1. Parameterization of the attosecond pulse

The few-cycle attosecond pulses used in this paper are chosen to have parameters similar to those produced experimentally in [1]: they have a frequency $\omega = 36$ eV and a duration of one or two optical cycles, resulting in a total duration of about 114.88 as for the one-cycle pulse and about 229.76 as for the two-cycle pulse. In contrast to the experimental attosecond pulses however, ours are assumed to have much higher peak intensities: $I = 1–2$ PW cm$^{-2}$. Such
The CEP \( \varphi \) (deg) dependence of the vector potential \( A(t) \) defined in equation (15) for an attosecond pulse with carrier frequency \( \omega = 36 \text{ eV} \) and two temporal durations: (a) one-cycle pulse and (b) two-cycle pulse. In both cases, the peak pulse intensity is \( I = 1 \text{ PW/cm}^{-2} \).

The explicit form of the linearly polarized attosecond pulse is defined by its vector potential \( A(t) \) and its electric field \( E(t) \) as follows:

\[
A(t) = A_0 f(t) \sin(\omega t + \varphi) e_z, \quad E(t) = -\frac{\partial}{\partial t} A(t),
\]

where \( \varphi \) is the CEP and \( f(t) \) is the pulse envelope,

\[
f(t) = \cos^2\left(\pi t / T\right) \quad \text{for} \ -T/2 \leq t \leq T/2,
\]

where \( T \) is the total pulse duration (chosen to be equal to one or two optical periods). The peak intensity of the pulse is given by \( I = E_0^2 \), where \( E_0 \equiv A_0 \omega \). In figure 1, we plot the vector potential \( A(t) \) defined in equation (15) for two temporal durations: a one-cycle pulse and a two-cycle pulse. The CEP dependence of each of these few-cycle attosecond pulses is clearly visible.

The carrier frequency of our pulses, \( \omega = 36 \text{ eV} \), is ideal for the investigation of the asymmetry in the ionization of electrons parallel or antiparallel to the \( z \)-axis in the vicinity of doubly excited autoionizing states. As predicted both numerically and theoretically for the hydrogen atom [19, 28], such asymmetries originate predominantly from interference of first- and second-order transition amplitudes for electron ionization. Such interference is significant if the energy bandwidth of the pulse is large and if the pulse intensity is sufficient for ensuring that the second-order amplitude is comparable in magnitude to that of the first-order amplitude in the energy region over which they overlap. The ionization threshold for the He atom is
approximately 24.6 eV, so that (for a long, essentially monochromatic pulse) electrons ionized by absorbing one or two photons should have kinetic energies of 11.4 and 47.4 eV, respectively. Owing to the energy–time uncertainty relation, i.e. $\Delta E \Delta t \gtrsim \hbar$, the bandwidths of our few-cycle pulses are such that there is overlap of these two amplitudes for energies in between those two energies. For the He atom, the doubly excited states converging to the He$^+(n = 2)$ threshold occur in the single-electron ionization spectrum from about 33.3 eV (i.e. the energy of the $2s^2(^1S^o)$ doubly excited state [71]) to the threshold at 40.8 eV, which is precisely the energy region over which we expect the one- and two-photon absorption amplitudes to overlap.

3.2. Overview of the single ionization of He by a few-cycle, intense attosecond pulse

Figure 2 presents the radial probability distribution of the two-electron wave packet (2) (with the ground-state component subtracted) integrated over the angular variables $(\hat{r}_1, \hat{r}_2)$ immediately after the end of the attosecond pulse for a peak pulse intensity of 2 PW cm$^{-2}$ and a CEP of $\pi/2$. The two-electron distributions in the radial variables $(r_1, r_2)$ are shown for two attosecond pulse durations: (a) one-cycle pulse; (b) two-cycle pulse. As expected, for the longer pulse the radial extent of the two-electron wave packet is roughly twice as large. Also, for the case of a two-cycle pulse, figure 2(b) shows ‘ring’ structures which are most likely evidence for interference between ionization amplitudes (including for double ionization) from different half-cycles of the pulse electric field; the one-cycle case in figure 2(a) does not show any ‘ring’ structure on the same scale. We note that no CEP effect can be seen in comparisons of the radial probability distributions of the two-electron wave packets in figure 2 with those for different values of the CEP (not shown). Nevertheless, as shown later, the CEP effect does appear in the differential probabilities (i.e. after projection of the wave packets onto field-free states).

Consider now the probabilities for electron ionization parallel ($\theta = 0$) and anti-parallel ($\theta = \pi$) to the z-axis (the direction of linear polarization of the attosecond pulse) that result from projecting the wave packets shown in figure 2 onto either the correlated $J$-matrix (CJM)
field-free states or the uncorrelated Sturmian–Coulomb (USC) field-free states. The results for both one-cycle and two-cycle pulses are shown in figures 3(a) and (b), respectively. One sees immediately in these figures that the projections onto the CJM and USC field-free states are in very good agreement in both directions along the $z$-axis with the exception of the energy region at which doubly excited states occur. In fact, proper treatment of two-electron correlation effects is essential in this energy region. Moreover, one sees also that there is very significant asymmetry between the probabilities for ionization along the directions $\theta = 0, \pi$. The asymmetry in the USC results is similar to that found for ionization of the H atom by an intense, few-cycle attosecond pulse [19, 28]. However, the asymmetry in the CJM results becomes much larger in the vicinity of the doubly excited states, as we shall discuss in the next subsection.

In our calculations, three ionization channels contribute significantly to the results shown in figure 3. Even for a peak pulse intensity of $2\text{ PW cm}^{-2}$, it is useful to interpret the non-perturbative TDSE results shown in figure 3 in terms of the key contributing perturbation theory terms (in the electron–laser pulse interaction) [28]. By electric dipole selection rules, the first-order transition amplitude produces final states with $^1P_o$ symmetry, while the second-order transition amplitude produces final states with either $^1S_e$ or $^1D_e$ symmetry. Owing to the different parities of the first- and second-order transition amplitudes, they interfere constructively on one side of the $z$-axis and destructively on the other. The third-order transition amplitude produces final states with either $^1P_o$ or $^1F_o$ symmetry. These are included in our calculations, although their contributions are small. (The largest contributions are for the third-order process in which two photons are absorbed and one is emitted leading to a $^1P_o$ final state, which combines with the first-order transition amplitude to interfere with the second-order terms.) We emphasize that in our numerical calculations we have used four values (0–3) of the total angular momentum.
L and our two-electron TDSE wave packet solution describes all orders of perturbation theory involving these terms.

In order to view in greater detail the electron correlation effects in the energy region of the doubly excited states, we show in figure 4 the energy region of the 2s2p(1P0) and 2p2(1D0) autoionizing states, which are populated, respectively, by the first- and second-order transition amplitudes (predominantly). In contrast to figure 3, we present the results for single ionization in the directions θ = 0, π separately (in figures 4(a) and (b), respectively) in order to also include the CEP dependence in each figure (i.e. the results for ϕ = 0, π/2). One sees immediately that the CJM results are much larger than the USC results. Moreover, the CJM results show Fano profiles [72], whereas the USC results are quite flat. These doubly excited state features appear in the CJM results owing to the electron correlation of the broadband distribution of ionized electron states with the electron in the residual He+ ion. We also see that the variation with CEP is substantial. Moreover, the line profiles in figures 4(a) and (b) have quite different magnitudes, which implies that their asymmetry (i.e. their absolute difference) will be large. Next we focus on these asymmetries in the region of autoionizing states.

3.3. Asymmetries in few-cycle attosecond pulse ionization of He in the vicinity of doubly excited states

The differential probability densities for few-cycle attosecond pulse ionization of He with emission of electrons either parallel (θ = 0) or antiparallel (θ = π) to the z-axis were shown in the previous section to differ significantly throughout the range of ionized electron energies displayed in figure 3, i.e. from threshold to 70 eV above. We consider now the energy dependence of the asymmetry, $\Delta P(E, \varphi)$ (defined by equation (13) with $\tilde{\Gamma} \equiv 1s$), for a fixed
Figure 5. Asymmetries $\Delta P(E, \varphi)$ (cf equation (13)) for attosecond pulse ionization of He to the ion ground state, $\text{He}^+(1s)$, with the ionized electron having a kinetic energy $E$ between 35 and 36 eV. The attosecond pulses all have a peak intensity of 2 PW cm$^{-2}$, but two different CEPs, $\varphi = 0, \pi/2$, and two pulse durations: (a) one-cycle pulse and (b) two-cycle pulse. Two autoionizing resonances having opposite parity are revealed in this energy region.

CEP. Note first that over the entire range of energies considered, i.e. from threshold to 70 eV above, we found that there are no significant differences between the asymmetries $\Delta P(E, \varphi)$ calculated by projecting onto CJM or USC states except for ionized electron energies in the range of 33–55 eV, where interactions with doubly excited states lead to major differences between the CJM and USC results. It is the behavior of the asymmetry $\Delta P(E, \varphi)$ in the vicinity of the lowest-energy doubly excited states that is our focus here.

In figure 5, we present the asymmetries $\Delta P(E, \varphi)$ (cf equation (13)) for ionized electron kinetic energies in the same energy interval as in figure 4: $35 \text{ eV} \leq E \leq 36 \text{ eV}$. For the two-cycle pulse case shown in figure 5(b), $\Delta P(E, \varphi)$ is equal to the difference of the differential probability densities, $P_{1s}(E, \theta, \varphi)$, for $\theta = 0$ and $\pi$ shown in figures 4(a) and (b), respectively, for each of the two values of the CEP $\varphi$. The corresponding asymmetries $\Delta P(E, \varphi)$ for the one-cycle case are shown in figure 5(a). Comparing the one-cycle and two-cycle cases in figures 5(a) and (b), one sees that the asymmetries have the same sign. For the case of a CEP $\varphi = \pi/2$, the origin of this similar behavior is easily understood: as shown in figures 1(a) and (b), the peak values of the vector potentials for the one-cycle and two-cycle pulse cases are both positive. As expected, the asymmetries for the projections onto USC field-free states are flat over the energy interval shown, with the results for a CEP $\varphi = \pi/2$ of larger magnitude than for a CEP $\varphi = 0$ owing to the greater asymmetry in the vector potential field (cf figure 1) in the former case. The projections onto CJM field-free states show the influence of the $2s2p(1P^o)$ and $2p^2(1D^o)$ autoionizing states, with the former originating predominantly from the first order interaction between the laser pulse and the He atom and the latter originating from the second order interaction. Remarkably, the influences of these autoionization states on the asymmetry are quite different for the CEPs of $\varphi = \pi/2$ and 0, with the latter having a smaller magnitude.
In order to provide a broader view of the combined electron correlation and few-cycle pulse-induced asymmetry effects, we display in figure 6 the asymmetry $\Delta P(E, \varphi)$ over a much wider interval of ionized electron energies, $32 \text{ eV} \leq E \leq 42 \text{ eV}$, containing the doubly excited states of He converging to the He$^+(n = 2)$ threshold. Comparing the one-cycle and two-cycle cases in figures 6(a) and (b), we see that the similar behavior observed in figure 5 is typical of the entire spectral range of the doubly excited states in figure 6. We note also that there is excellent agreement between the resonance positions shown in figure 6 and predictions in the literature (see, e.g., [71] and references therein). This may be viewed as a confirmation of the numerical accuracy of the present calculations. The appearance of odd and even parity autoionizing resonances proves the occurrence of interference between states of different total angular momenta (i.e. $L = 0, 1$ and 2). Despite the interferences between transitions to states of different angular momenta and parities, the signatures of many of the autoionizing resonances remain visible and are identified.

3.4. Energy-integrated asymmetries in few-cycle attosecond pulse ionization of He

In figures 7 and 8, we plot the energy-integrated asymmetries $\Delta P(\varphi)$ (cf equations (12) and (14)) for the cases of a one-cycle pulse and a two-cycle pulse, respectively. (Note that in evaluating the integral in equation (12) the limits of integration were chosen as 0.1 and 100 eV.) As expected, the integrated asymmetries for the one-cycle and two-cycle pulse cases have similar signs (owing to the same signs of the peak values of their respective vector potentials, shown in figure 1). Moreover, the shorter duration of the one-cycle pulse leads to a larger magnitude of the asymmetry than for the case of the two-cycle pulse. One observes that there do exist differences between the energy-integrated asymmetries obtained from CJM and USC projections, but these differences are small because the magnitudes of the differential probabilities in the energy region of the doubly excited state resonances are small compared to
Figure 7. CEP dependence of the energy-integrated asymmetries $\Delta P(\varphi)$ (cf equations (12) and (14)) for the case of a one-cycle pulse and two values of the peak pulse intensity: (a) 1 PW cm$^{-2}$ and (b) 2 PW cm$^{-2}$.

Figure 8. The same as figure 7 but for the case of a two-cycle pulse.

those at lower energies. However, the differences between these two projections become greater as the peak pulse intensity increases.

The dependence of the energy-integrated asymmetry $\Delta P(\varphi)$ on the CEP $\varphi$ has a concave shape in both the one-cycle pulse case (cf figure 7) and the two-cycle pulse case (cf figure 8). Also, the peak magnitude of the energy-integrated asymmetry for the one-cycle pulse is roughly twice as large as that for the two-cycle pulse, which is consistent with the greater asymmetry in the vector potential of the one-cycle pulse as compared to that of the two-cycle pulse (cf figure 1) as well as with the fact that the asymmetry vanishes in the case of a monochromatic pulse of infinitely many cycles. Moreover, whereas the peak magnitude of the energy-integrated
Table 1. Dynamical parameters $|K|$, $\Theta_K$ obtained through a nonlinear fit (using a statistics package in Maple) of equation (17) to our one- and two-cycle CJM data for pulse intensities of 1 and 2 PW cm$^{-2}$.

<table>
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<tr>
<th>Pulse parameters:</th>
<th>115 as/1 PW</th>
<th>115 as/2 PW</th>
<th>230 as/1 PW</th>
<th>230 as/2 PW</th>
</tr>
</thead>
<tbody>
<tr>
<td>$</td>
<td>K</td>
<td>$ (10$^{-2}$ au):</td>
<td>1.8588</td>
<td>5.2575</td>
</tr>
<tr>
<td>$\Theta_K$ (deg):</td>
<td>84.55</td>
<td>84.70</td>
<td>64.72</td>
<td>64.91</td>
</tr>
</tbody>
</table>

Table 2. The same as table 1 but for the USC data.

<table>
<thead>
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<th>Pulse parameters:</th>
<th>115 as/1 PW</th>
<th>115 as/2 PW</th>
<th>230 as/1 PW</th>
<th>230 as/2 PW</th>
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</thead>
<tbody>
<tr>
<td>$</td>
<td>K</td>
<td>$ (10$^{-2}$ au):</td>
<td>1.7915</td>
<td>5.0306</td>
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<tr>
<td>$\Theta_K$ (deg):</td>
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<td>85.42</td>
<td>66.08</td>
<td>66.35</td>
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</table>

asymmetry occurs at $\varphi = \pi/2$ in the case of the one-cycle pulse, it occurs at $\varphi = \pi/3$ in the case of the two-cycle pulse, indicating a complex interplay between the amplitudes for ionization in different cycles of the few-cycle pulse.

For the few-cycle attosecond pulses employed in our numerical calculations, which have a carrier frequency of $\omega = 36$ eV and peak pulse intensities of 1–2 PW cm$^{-2}$, the strength of the pulse electric field is sufficiently weak (compared with the Coulomb field experienced by the electrons) that time-dependent perturbation theory in powers of the pulse electric field may be employed [28]. For a linearly polarized pulse, Pronin et al. [28] have established that the CEP dependence of the energy-integrated asymmetry has the following form (see equations (29), (74) and (75) of [28]):

$$\Delta P(\varphi) = |K| \cos(\varphi - \Theta_K),$$

where $K \equiv |K| \exp(i \Theta_K)$ is a complex dynamical parameter that does not depend on the CEP $\varphi$. Using our numerical data for the energy-integrated asymmetries, we have performed a nonlinear fit (using a statistics package in Maple) of equation (17) to both our CJM and USC data in order to determine those two dynamical parameters. The values of $|K|$ and $\Theta_K$ obtained from these fits are given in tables 1 and 2. One sees that the complex phase $\Theta_K$ is much less sensitive to electron correlation effects than the magnitude $|K|$ for all laser pulse parameters. Moreover, the agreement between the CJM and USC dynamical magnitudes $|K|$ is better for the 115 as pulse case than for the 230 as pulse case. Finally, we see that the magnitudes $|K|$ change dramatically for different pulse intensities, unlike the case of the complex phase $\Theta_K$.

Comparisons between the predictions for the energy-integrated asymmetries using the perturbation theory result of equation (17) with our numerical CJM and USC results are given in figures 7 and 8. In the case of a two-cycle pulse (cf figure 8), one sees that there is essentially exact agreement between our numerical CJM and USC predictions for the CEP dependence of the attosecond pulse asymmetries and the predictions of time-dependent perturbation theory [28] (which include terms up to second order in the attosecond pulse). In other words, the CEP dependence predicted by perturbation theory (cf equation (17)) is accurate regardless of whether the perturbation theory parameters are fit to the numerical CJM or USC results. In the case of a one-cycle pulse however, the agreement is fairly good, but not so exact as in the
two-cycle case (cf figure 7). In particular, there are significant discrepancies in the magnitudes of the maxima at a CEP of $\pi/2$. We suspect that these disagreements of our numerical results with the perturbation theory formula (17) for the case of a one-cycle pulse originate from the contributions of three-photon processes in our numerical results that are not taken into account in the perturbation theory formula. For a monochromatic laser field, electrons ionized by absorbing three photons should appear with a kinetic energy of 83.4 eV. The energy over which we have integrated the differential probability densities (i.e. from 0.1 to 100 eV) includes this energy. Moreover, three-photon processes in which two photons are absorbed and one photon is emitted should occur in the energy region for one-photon ionization. The much larger bandwidth of a one-cycle pulse (as compared to that for a two-cycle pulse) results in much more significant overlap of the amplitudes for such three-photon processes with the amplitudes for one- and two-photon processes (than in the case of a two-cycle pulse). Conversely, the excellent agreement of our two-cycle pulse results with the perturbation theory formula (17) is an indication of the minimal role played by the amplitudes for three-photon processes in that case.

The perturbation theory analysis of attosecond photoionization provides many symmetry relations obeyed by the ionization spectra [28]. In the case of linearly polarized light, both the differential probability density and the energy-integrated probability are expected to be invariant under the $\phi \leftrightarrow \theta$ transformation. Also, one has that $P(E, \theta, \phi) = P(E, \pi - \theta, \pi + \phi)$. Thus, one expects for any given energy $E$ the following symmetry relations: $P(E, 0, \phi) = P(E, \pi, \pi)$ and $P(E, 0, \pi) = P(E, \pi, 0)$. Applying these symmetry relations to the energy-integrated asymmetry, one expects that $\Delta P(\phi = 0) = -\Delta P(\phi = \pi)$, in agreement with the CEP dependence predicted by equation (17). Our CJM and USC results confirm all these predictions, in particular, the latter one, as may be seen in figures 7 and 8.

Note, finally, that all our results shown here have focused on the ionization of electrons along the $z$-axis and hence the question arises of whether or not such asymmetries occur if one carries out an angular integration over the forward and backward hemispheres. We found that there is no major change in the expected results owing to the fact that for the case of linear polarization forward and backward electron ejection along the polarization axis provides the dominant contribution among all angles in either the forward or backward hemisphere.

4. Summary and conclusions

In summary, we have solved the fully dimensional TDSE for He, the prototypical two-active-electron system interacting with an intense, few-cycle attosecond pulse. The differential probability distributions are then obtained by projecting the time-dependent wave packet solution of the TDSE onto two field-free sets of states, the correlated $J$-matrix (CJM) and the uncorrelated Sturmian–Coulomb (USC) states. For ionized electron energies below the region of He doubly excited states we find little difference in the results for these two different field-free states. However, for ionized electron energies in the region of the He doubly excited states, we find significant differences, with the projections onto the CJM states revealing the signatures of the doubly excited states on the differential probability densities for single-electron ionization. Experimentally, since we treat the case when the ion is left in its ground state, He$^+(1s)$, a measurement of the ionized electron energy spectrum in the vicinity of the autoionizing states should reveal the effects of those doubly excited states predicted by our CJM results.

Such clear evidence of electron correlations is remarkable for the ultrashort few-cycle pulses we employ. Indeed, our one-cycle pulses have a total duration of 115 as, which is much
shorter than the lifetime of the $2s2p(^1P^o)$ doubly excited state. Moreover, the energy bandwidths of our pulses are of the order of tens of eV, which is far broader than the widths of the doubly excited states we observe in our predicted single-ionization spectra. We interpret these facts as follows: our ultrashort attosecond pulses produce a broad energy spectrum of singly excited electron states; in the energy region of the doubly excited states, electron correlations between the continuum electrons and the doubly excited states result in the observed doubly excited state signatures in the singly ionized electron spectrum.

We have also investigated the CEP dependence of the asymmetries in the spectrum of continuum electrons ionized with momenta along the positive and negative directions of the $z$-axis, the axis of linear polarization of the attosecond pulses. We found excellent agreement of our numerical predictions with the second order, time-dependent perturbation theory analysis of attosecond photoionization in [28].

Acknowledgments

We gratefully acknowledge very useful discussions with M V Frolov, N L Manakov and B Piraux. This work was supported in part by the US Department of Energy, Office of Science, Division of Chemical Sciences, Geosciences, and Biosciences, under grant number DE-FG03-96ER14646, by the Laboratory for Laser Energetics at the University of Rochester and by the National Natural Science Foundation of China under grant numbers 11174016, 10974007, 11121091 and 11134001. The computational results were obtained using the Merritt computational facility at the Holland Computing Center of the University of Nebraska, the US National Science Foundation TeraGrid computational resources on the National Institute for Computational Sciences’ Kraken supercomputer under grant numbers TG-PHY-100052, TG-PHY-120003, TG-PHY-110003, and TG-PHY-110009, and by the computer cluster ‘MESO’ in the State Key Laboratory for Mesoscopic Physics at Peking University.

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