Generation of attosecond electron beams in relativistic ionization by short laser pulses

F Cajiao Vélez, J Z Kamiński and K Krajewska

Institute of Theoretical Physics, Faculty of Physics, University of Warsaw, Pasteura 5, 02-093 Warsaw, Poland

E-mail: Katarzyna.Krajewska@fuw.edu.pl

Received 2 May 2017, revised 29 November 2017
Accepted for publication 8 December 2017
Published 1 February 2018

Abstract

Ionization by relativistically intense short laser pulses is studied in the framework of strong-field quantum electrodynamics. Distinctive patterns are found in the energy probability distributions of photoelectrons, which are sensitive to the properties of a driving laser field. It is demonstrated that these electrons are generated in the form of solitary attosecond wave packets. This is particularly important in light of various applications of attosecond electron beams such as in ultrafast electron diffraction and crystallography, or in time-resolved electron microscopy of physical, chemical, and biological processes. We also show that, for intense laser pulses, high-energy ionization takes place in narrow regions surrounding the momentum spiral, the exact form of which is determined by the shape of a driving pulse. The self-intersections of the spiral define the momenta for which the interference patterns in the energy distributions of photoelectrons are observed. Furthermore, these interference regions lead to the synthesis of single-electron wave packets characterized by coherent double-hump structures.

Keywords: intense laser pulses, relativistic ionization, supercontinuum, attosecond electron pulses

((Some figures may appear in colour only in the online journal)

1. Introduction

In recent years, the synthesis of electron wave packets of short duration has attracted a lot of attention due to its applications in ultrafast electron diffraction, crystallography, and microscopy [1–4]. It has been shown that femtosecond electron pulses can be used to image complex molecular, biological, and crystalline structures with very short temporal resolution. Therefore, it has become possible to observe transient molecular structures in the course of chemical reactions [5–9], gather an insight into melting and heating processes [10–12], or to observe phase transitions in crystalline and polycrystalline materials [13, 14].

Short electron wave packets can be produced by different methods. The traditional ones make use of the photoelectric effect by shining laser pulses of short duration upon flat photocathodes [15] or sharp metallic tips (see [16] and references therein). Other techniques include electron emission in intense plasmon fields [16–20] and photoemission from supercooled atoms in optical traps [21–23]. The low-energy photoelectrons obtained by these methods appear with velocity distributions that depend on the geometry of the source [15]. For applications in microscopy or diffraction, those electrons need to be accelerated up to appropriate kinetic energies [24]. During the acceleration step and posterior free propagation, the non-relativistic wave packets spread in time. Furthermore, if the short pulses comprise a large number of photoelectrons, an additional broadening is expected as a consequence of Coulomb repulsion [25, 26]. Thus, in order to minimize this broadening, synthesis of ultrashort wave packets containing a single electron has been considered [3, 15, 24, 27–30].

The shortest single-electron pulses with a full width at half maximum duration of 28 fs have been demonstrated recently [31]. Moreover, various proposals for further compression of these pulses to attosecond durations have been put forward [27, 28, 32–35]. At this point, it is also important to note that theoretical investigations of scattering processes employing attosecond electron pulses have confirmed their ability to image electronic motions in target atoms and molecules [36–39].
Yet, another proposal for producing attosecond electron pulses has been introduced in [40]. In this paper, we have shown that very short electron bunches, with energies of few keV, can be produced by the interaction of hydrogen-like ions with intense circularly polarized laser fields. In this case, a broad structure located at the high-energy portion of the photoelectron spectrum is formed (see also [41]). This structure, the so-called supercontinuum, is characterized by the absence of multiphoton interferences. Moreover, it does not present significant fluctuations of probability in the range of tens to hundreds single-photon energies and, according to the space–time analysis, can be used to obtain electron pulses with attosecond duration.

It is the aim of this paper to further analyze the properties of the supercontinuum in photoionization and its application to the generation of attosecond electron wave packets. First, we show that the photoelectron energy spectra can exhibit, in general, many well-defined interference-free structures at different photoelectron kinetic energies. Next, we show that these structures can be used to synthesize attosecond electron pulses. As we demonstrate, the time duration of the resulting electron pulses can be shortened by increasing the intensity of the laser field which drives ionization.

In contrast to our previous works [40, 41], we consider here ionization stimulated by low-frequency laser fields. This has been motivated by the fact that such fields, with high intensities and short durations, will soon be available in laser facilities such as Extreme Light Infrastructure (ELI) [42] or Exawatt Center for Extreme Light Studies (XCELS) [43]. Moreover, it appears beneficial to use low-frequency incident laser fields in the context of electron pulse generation. The point is that, if we fix the laser field intensity, the lower its frequency the more energetic photoelectrons will be detected (since the ponderomotive energy of electron oscillations in a laser field increases). This, in turn, will lead to the synthesis of shorter electron pulses.

We would like to point out that the synthesis of attosecond electron pulses from the above-mentioned high-energy supercontinuum may have at least two advantages. First, the detected photoelectrons have velocities appropriate for ultrafast electron microscopy and diffraction, which makes their further acceleration unnecessary. Second, those photoelectrons are observed within a narrow angular window and, depending on the actual size of the final electron beam, additional collimation methods might be less needed. Note, however, that ultrashort electron wave packets have large energy bandwidths, as a direct consequence of the uncertainty principle. It is, therefore, expected that nonrelativistic short electron pulses spread fast in time during free propagation. This is not the case for wave packets synthesized with relativistic kinetic energies (close to MeV), as their natural spreading is generally slower. We show here that very energetic photoelectrons can actually be employed by employing laser fields of larger intensity. Nevertheless, for electron microscopy applications, standard compression techniques can also be applied [27, 28, 44–48].

This paper is organized as follows. In section 2, for convenience of the reader, we present the formulas of the photoionization probability distributions under the scope of the relativistic strong-field approximation (RSFA) and the plane-wave front approximation. The respective derivations were initially presented in [40]. While section 3 relates to the energy spectra of photoelectrons and the formation of the supercontinuum, section 4 is dedicated to the analysis of the space–time probability distributions and the formation of attosecond electron wave packets. Also in section 4, the validity of the plane-wave front approximation for the driving laser pulse is tested for our calculations. In section 5, we define the so-called momentum spiral and show that, for intense laser pulses, high-energy ionization occurs mostly for momenta closely surrounding the spiral. Moreover, for electron momenta from the vicinity of self-intersections of the spiral, the interference patterns in the probability distribution of ionization occur. We demonstrate that such interference patterns lead to a double-hump structure in the space–time distribution of the resulting electron wave packets. This will be discussed in section 6. Finally, in section 7 we summarize our results and outline the perspectives for further investigations.

2. Theoretical formulation

Consider a hydrogen-like ion interacting with a relativistically strong laser pulse. The probability amplitude of ionization from the initial bound state of energy $E_b$, $\Psi_i(x)$, to the final scattering state, $\Psi_f(x)$, is given as [40, 41, 49]

$$A_0 = -i \int d^4x e^{-iE_b(x)/\hbar} \Psi_i(x) e^{iA_R(x)} \Psi_f(x),$$

where the four-vector $A_R(x)$ represents the electromagnetic potential describing the laser field and $e < 0$ is the electron charge. While equation (1) is exact and $\Psi_i(x)$ can be derived analytically for the Coulomb potential (see, e.g., [50]), $\Psi_f(x)$ can only be determined numerically for laser fields of moderate intensities. For this reason, we shall use now the RSFA [40], in which the exact scattering state $\Psi_f(x)$ is treated using the lowest-order Born approximation. This approximation is justified when the kinetic energy of photoelectrons is much larger than the ionization potential of the initial bound state, i.e., when $\sqrt{(m_e c^2)^2 + (cp)^2} - m_e c^2 \gg m_e c^2 - E_b$, as discussed in [40, 41]. Note that this condition is very well satisfied for high-energy photoelectrons, which are the main focus of our paper.

In the RSFA, the spin-fixed probability amplitude of ionization (1), now denoted as $A_{\lambda,\gamma}(p)$, takes the form

$$A_{\lambda,\gamma}(p) = -i \int \frac{d^3q}{(2\pi)^3} \int d^3x e^{-i\vec{q}\cdot\vec{p}} \Psi_i(x) \times e^{iA_R(x)} \Psi_f(q),$$

where the exact scattering state has been replaced by the Volkov solution [51] (i.e., the solution of the Dirac equation
coupled to the laser field). Here, \( \psi_{p}^{(+)}(x) \) is the Volkov solution describing the electron with an electromagnetic potential \( A_{\mathbf{r}}(x) \) and a spin \( \lambda = \pm \) \( \mathring{\psi}(q) \) is the Fourier transform of the bound state \( \mathring{\psi}(x) \) and \( \lambda_{s} \) is the initial electron spin. Note that, in equation (2), we have introduced \( q = (\omega, \mathbf{q}) = (E_{0}/c, \mathbf{q}) \), which is not a four-vector but does not transform properly under Lorentz transformations. Nevertheless, it will help us to simplify our further notation.

At this point, we stress that the Born approximation is essentially the only approximation in our theory. Below, in order to use the analytical form of the Volkov solution, we will neglect the longitudinal components of the laser field. This, however, is justified in the current case as the entire dynamics of high-energy electrons in the laser pulse takes place close to the symmetry axis of the laser focus (see, the discussion in section 4.4).

Our calculations are carried out in the velocity gauge and the laser field is described using the plane-wave front approximation. In this case, the electromagnetic potential \( A_{\mathbf{r}}(x) \) can be written as

\[
A_{\mathbf{r}}(x) = A_{0}[\xi_{1} f_{1}(k \cdot x) + \xi_{2} f_{2}(k \cdot x)],
\]

where \( k = k_{0}n = k_{0}(1, \mathbf{n}) \) is the wave four-vector and \( k_{0} = \omega/c \). In our notation, the unit vector \( \mathbf{n} \) represents the direction of propagation of the laser pulse, \( \omega = 2\pi/T_{p} \) is its fundamental frequency, whereas \( T_{p} \) represents its duration. Additionally, \( \xi_{j} \equiv (0, \xi_{j}) \) \( (j = 1, 2) \) are two real and normalized polarization four-vectors perpendicular to the laser field propagation direction (i.e., \( k \cdot \xi_{j} = -k \cdot \xi_{j} = 0 \)). In equation (3), \( f_{j}(\phi) \) represent two real shape functions with continuous second derivatives which vanish for \( \phi \lt 0 \) and \( \phi \gt 2\pi \).

We recall that the Volkov solution for an electron, normalized in the volume \( V \), equals [51–53]

\[
\psi_{p}^{(+)}(x) = \sqrt{\frac{m_{e}c^{2}}{VE_{p}}} \left( 1 + \frac{m_{e}c\mu}{2p \cdot k} [f_{1}(k \cdot x) \xi_{1} \xi_{1} k + f_{2}(k \cdot x) \xi_{2} \xi_{2} k] \right) e^{-i\mathring{G}_{\mu}(\mathbf{u}_{p}^{(+)})},
\]

where \( m_{e} \) is the electron mass, \( p = (E_{p}/c, \mathbf{p}) \) is its asymptotic on-shell four-momentum, whereas

\[
S_{p}^{(+)}(x) = \mathring{G}_{\mu}(\mathbf{u}_{p}^{(+)}) = \int_{0}^{k_{x}} d\phi \left[ -\frac{m_{e}c\mu}{p \cdot k} (\xi_{1} \cdot p\mu f_{1}(\phi) + \xi_{2} \cdot p\mu f_{2}(\phi)) + \frac{m_{e}c\mu^{2}}{2p \cdot k} [f_{1}^{(2)}(\phi) + f_{2}^{(2)}(\phi)] \right] \]

Moreover, \( \mu = |eA_{0}|/m_{e}c \) denotes the normalized amplitude of the vector potential (3) whereas \( u_{p}^{(+)}(x) \) is the free-electron bispinor [50]. Using these expressions, we derive that the probability amplitude of ionization under the RSFA (2) becomes

\[
\mathcal{A}_{\lambda_{l},\lambda}(p) = \int \frac{d^{3}q}{(2\pi)^{3}} \int d^{3}x e^{i\mathring{G}_{\mu}^{(+)}}(x) \mathcal{D}(p, \lambda; \lambda_{s}) \mathcal{M}_{\lambda_{l},\lambda}(k \cdot x),
\]

where

\[
\mathcal{M}_{\lambda_{l},\lambda}(k \cdot x) = \frac{m_{e}c^{2}}{VE_{p}} \left[ f_{1}(k \cdot x)B_{p\lambda_{l},\lambda}^{1,0}(q) + f_{2}(k \cdot x)B_{p\lambda_{l},\lambda}^{0,1}(q) \right] - \frac{m_{e}c\mu}{2p \cdot n} \left[ f_{1}^{(2)}(k \cdot x) + f_{2}^{(2)}(k \cdot x) \right] B_{p\lambda_{l},\lambda}(q).
\]

To simplify our notation, we have introduced the following functions expressed in terms of the Fourier transform of the initial ground state, \( \mathring{\psi}(q) \),

\[
B_{p\lambda_{l},\lambda}^{1,0}(q) = \mathring{\psi}_{\lambda_{l}}^{(+)}(q),
\]

\[
B_{p\lambda_{l},\lambda}^{0,1}(q) = \mathring{\psi}_{\lambda}^{(+)}(q),
\]

\[
B_{p\lambda_{l},\lambda}(q) = \mathring{\psi}_{\lambda_{l}}^{(+)}(q).
\]

In the next step, we define the so-called laser-dressed four-momentum of the electron, \( p_{l} \),

\[
\mathcal{D}(p, \lambda; \lambda_{s}) = \int_{0}^{\phi_{p}} d\phi' \left[ -\frac{m_{e}c\mu}{p \cdot k} (\xi_{1} \cdot p(f_{1}(\phi') - (f_{1})) + \xi_{2} \cdot p(f_{2}(\phi')) - (f_{2})) + \frac{(m_{e}c\mu)^{2}}{2p \cdot k} [f_{1}^{(2)}(\phi') + f_{2}^{(2)}(\phi')] \right],
\]

Now, it is useful to introduce the light-cone coordinates in equation (6), namely, \( x = x^{0} - n \cdot x, x^{+} = \frac{1}{2}(x^{0} + n \cdot x) \), and \( x^{-} = x - (n \cdot x)n \). Since the pulse phase \( k \cdot x = k_{0}x^{-} \), most of the integrals in equation (6) become trivial. The only nontrivial integral over \( x^{-} \) is performed with the help of the Fourier expansions [54, 55],

\[
[f_{1}(\phi)]^{t} \exp[iG_{p}(\phi)] = \sum_{N=-\infty}^{\infty} G_{N}^{(i\phi)} e^{-iN\phi},
\]

\[
[f_{2}(\phi)]^{t} \exp[iG_{p}(\phi)] = \sum_{N=-\infty}^{\infty} G_{N}^{(i\phi)} e^{-iN\phi},
\]

where \( j = 1, 2 \). Hence, the spin-resolved probability amplitude of ionization can be represented as an infinite sum,

\[
\mathcal{A}_{\lambda_{l},\lambda}(p) = \frac{m_{e}c^{2}}{VE_{p}} \mathcal{D}(p, \lambda; \lambda_{s}).
\]
where
\[
\mathcal{D}(\mathbf{p}; \lambda) = \sum_{N=-\infty}^{\infty} \frac{e^{2\pi i \mathbf{p} \cdot \mathbf{q} - q \cdot \mathbf{N} \lambda / \mathbf{q} - \mathbf{p} \cdot \mathbf{N} \lambda}}{i(\mathbf{p} \cdot \mathbf{q})^2 - N^2 \lambda^2} \times \left\{ G_{N}^{(1,0)} B_{\lambda \lambda}^{(1,0)}(Q) + G_{N}^{(0,1)} B_{\lambda \lambda}^{(0,1)}(Q) \right\} - \frac{m e c^2}{2p_n} \left[ G_{N}^{(1,0)} B_{\lambda \lambda}^{(2,0)}(Q) + G_{N}^{(0,1)} B_{\lambda \lambda}^{(0,2)}(Q) \right]
\]
and \( Q = p + (q^0 - p^0)m \).

Finally, integrating \( |A_{\lambda \lambda}(p)|^2 \) over the density of final electron states, \( \mathcal{V} d^3p/(2\pi)^3 \), we obtain that the total probability of ionization equals
\[
P_{\text{ion}} = \frac{\mu^2 (m_e c)^3}{2(2\pi)^3} \sum_{\lambda \lambda = \pm} \int \frac{d^3p}{p^0} |\mathcal{D}(\mathbf{p}; \lambda)\lambda|^2.
\]

Here, the averaging over the initial and summation over the final electron spin degrees of freedom have been performed. Furthermore, the above equation allows us to define the spin-resolved triply differential probability distribution of ionization,
\[
\frac{d^3P(p; \lambda; \lambda)}{dE_p d^2\Omega_p} = \frac{\mu^2 (m_e c)^3}{(2\pi)^3} |\mathcal{D}(\mathbf{p}; \lambda; \lambda)|^2.
\]

For the purpose of our numerical illustrations, we introduce also the dimensionless distributions,
\[
\mathcal{P}_{\lambda \lambda}(p) = \alpha \frac{m_e c^2}{2} \sum_{\lambda \lambda = \pm} \frac{d^3P(p; \lambda; \lambda)}{dE_p d^2\Omega_p},
\]
and
\[
\mathcal{P}(p) = \frac{\alpha^2 m_e c^2}{2} \sum_{\lambda \lambda = \pm} \frac{d^3P(p; \lambda; \lambda)}{dE_p d^2\Omega_p},
\]
where \( \alpha = e^2/(4\pi\varepsilon_0 c) \) is the fine-structure constant. These are the probability distributions of ionization expressed in atomic units [40].

2.1. Laser pulse and hydrogen-like ion prototype

To illustrate the theory presented above, we consider the interaction of an intense and circularly polarized laser pulse with He\(^+\) ions. The latter are one-electron ions, with the atomic number \( Z = 2 \). Their ground state energy equals \( E_0 = m_e c^2 \sqrt{1 - (Z\alpha)^2} \) and, hence, the ionization potential, \( m_e c^2 - E_0 \), is roughly 54 eV. The corresponding ground state wave functions are [50]:
\[
\psi(x) = N_\lambda \rho^{\gamma - 1} e^{-m_e c^2 x / m_e c^2} \begin{bmatrix} 1 \\ \frac{1 - \gamma}{Z_0} \sin \theta e^{i\phi} \\ \frac{1 - \gamma}{Z_0} \cos \theta \end{bmatrix}
\]
for \( \lambda = + \), and
\[
\psi(x) = N_\lambda \rho^{\gamma - 1} e^{-m_e c^2 x / m_e c^2} \begin{bmatrix} 0 \\ \frac{1 - \gamma}{Z_0} \sin \theta e^{-i\phi} \\ \frac{1 - \gamma}{Z_0} \cos \theta \end{bmatrix}
\]
for \( \lambda = - \). Here, \( \gamma = E_0/(m_e c^2) \), \( N_\lambda = (2\pi c Z_0)^{1/2} / (4\pi e^2)^{1/2} \), \( \Gamma(x) \) is the Gamma function, and \((r, \theta, \phi)\) are the spherical coordinates of the position vector \( x \).

The laser pulse (3) is characterized by the shape functions \( f_j(\phi) \), for \( j = 1, 2 \), defined as
\[
f_j(\phi) = - \int_0^\phi d\phi F_j(\phi'),
\]
where
\[
F_j(\phi) = N_0 \sin^2 \left( \frac{\phi}{2} \right) \sin(N_\text{osc} \phi + \delta_j) \cos(\delta + \delta_j)
\]
for \( 0 < \phi < 2\pi \) and it is 0 otherwise. Here, \( N_\text{osc} \) is the number of cycles comprising the pulse, \( N_0 = \sqrt{3} N_\text{osc} \) is a normalization constant which guarantees that the average intensity of the field is independent of the number of cycles [40], whereas \( \delta \) and \( \delta_j \) determine the polarization properties of the pulse. Unless otherwise stated, in the following, we consider an isolated three-cycle Ti:Sapphire laser pulse, with the wavelength of 800 nm, the photon energy 1.5498 eV, the carrier laser frequency \( \omega_1 = 2.36 \times 10^{13} \) s\(^{-1} \), and pulse duration 8 fs. The numerical results refer to the averaged intensity of the laser pulse. The peak intensity is then 8/3 times larger. The laser pulse is right-handed circularly polarized (\( \delta_1 = 0, \delta_2 = \pi/2 \), and \( \delta_3 = \pi/4 \)) and propagates in the \( z \)-direction (\( \mathbf{e}_z \)) with the polarization vectors \( \mathbf{e}_1 = \mathbf{e}_z \) and \( \mathbf{e}_2 = \mathbf{e}_z \). Note that in our previous studies [40, 41], we have used the laser field corresponding to much shorter wavelength of 62 nm.

In figure 1, we present the trajectories of the tips of the electric field vector \( \mathbf{E}(\phi) \) (upper panel) and the vector potential \( A(\phi) \) (lower panel) in the \( xy \)-plane for the laser pulse described above. Both curves start and end up at the origin of coordinates and evolve counterclockwise. The parameter \( \mathbf{E}_\phi = m^2 c^3/|e| \) is the so-called Sauter–Schwinger critical field [56, 57], whereas \( A_\phi = m_e c^2/|e| \). Note that, for the laser field parameters considered in this paper, we have \( |\mathbf{E}(\phi)/\mathbf{E}_\phi| \ll 1 \). This implicates that, under current conditions, the electron–positron pair creation from vacuum is negligible [40].

2.2. Monte Carlo analysis

In section 2 we have defined the triply differential probability distribution of ionization of hydrogen-like ions interacting with short laser pulses (equation (15)). In this case, the initial ground state wave function is unperturbed at times prior to the interaction with the laser field, independently of its intensity. This allows us to analyze the photoionization of light ions (such as He\(^+\)) by intense laser pulses. In contrast, when the infinite plane-wave approximation is considered (see, e.g., [49, 58–60]), the RSFA is restricted to the photoionization of highly charged positive ions. This guarantees that the initial bound state is not heavily distorted by the action of the laser field.

Note also that, when a slowly varying envelope is used to model the driving laser pulse, the so-called Lambropoulos curve can play a role in the dynamics of photoionization [61].
Namely, it may happen that the hydrogen-like ion is fully ionized during the ramp-up part of the laser pulse, before the field acquires its maximum strength. In contrast, when the envelope varies rapidly, the field reaches values large enough to permit stabilization against ionization in very short time intervals. In this case, the target may survive undisturbed even up to the maximum field strength in the pulse [62].

As described above, our approach offers many advantages as compared to the case when the infinite plane-wave laser field is considered. However, it is still crucial to show that the unitarity of the problem is not violated while applying the approximations described in this paper. In other words, that the total probability of ionization (14) is always less than one. To do so, we calculate below $P_{\text{ion}}$ using the Monte Carlo method.

First, we rewrite equation (14) such that

$$P_{\text{ion}} = \frac{1}{\alpha^2 m_e c^2} \int_0^{2\pi} d\varphi \int_{-1}^1 d\cos \theta_p \int_{m_e c^2}^\infty dE_p \mathcal{P}(p),$$

where $\mathcal{P}(p)$ is defined by equation (17). To numerically evaluate the energy integral, its upper limit is substituted by a finite value, denoted as $m_e c^2 + E_{\text{max}}$. It is chosen such that the probability distribution is negligible for photoelectron final energies larger than $E_{\text{max}}$. In our calculations, we set $E_{\text{max}} = 30$ keV. Now, by introducing the following change of variables,

$$\varphi_p = 2\pi \xi_1, \cos \theta_p = 2\xi_2 - 1, E_p = m_e c^2 + E_{\text{max}} \xi_3,$$

the total probability of ionization becomes

$$P_{\text{ion}} = \frac{4\pi m_e c^2}{\alpha^2} \int_0^1 d\xi_1 \int_{-1}^1 d\xi_2 \int_{-1}^1 d\xi_3 \mathcal{P}(p),$$

where we integrate over a unit cube. This expression is now suitable to apply the Monte Carlo method with the uniformly distributed variables $\xi_i$ ($i = 1, 2, 3$) (see, e.g., [63, 64]).

In figure 2, we show the total probabilities of photoionization of He$^+$ ions by the laser pulse described in the previous section. The data are for different time-averaged intensities such that $I = 2 \times 10^{16}$ W cm$^{-2}$ (diamonds), $I = 5 \times 10^{16}$ W cm$^{-2}$ (squares), and $I = 1 \times 10^{17}$ W cm$^{-2}$ (stars), and for different number of field cycles, $N_{\text{osc}}$. The results were obtained from the Monte Carlo integration of equation (24) by considering no less than 10$^6$ sample points, with the estimated relative standard deviations smaller than 1%. One can clearly see that all calculated probabilities are less than one, which indicates the self-consistency of our treatment. With this in mind, we can study now differential probability distributions of ionization and their properties.

### 3. Energy probability distributions of ionization

As was mentioned before, the validity of the RSFA is restricted to the case when the kinetic energy of photoelectrons is much larger than the ionization potential of the parent...
Figure 3. Ionization probability distributions (17) for the azimuthal angle $\varphi_p = 0.5\pi$ and two polar angles, $\theta_p = 0.46\pi$ (upper panel) and $\theta_p = 0.5\pi$ (lower panel). The laser pulse parameters are the same as in figure 1.

Figure 4. Color mappings of ionization probability distributions $\mathcal{P}(p)$ for the azimuthal angle $\varphi_p = 0.5\pi$. The intermediate- and high-energy structures are shown in the upper and lower panels, respectively. The laser field parameters are the same as in figure 1.

Ion. Nevertheless, in this section we are going to present the angular-resolved ionization probability distributions for photoelectron kinetic energies ranging from 0 up to $10^5\omega_L$. It is, therefore, important to keep in mind that the results corresponding to the low-energy part of the spectra provide only a qualitative insight into the process.

In figure 3, we present the respective energy distributions of ionized electrons when measured at fixed angles. Such distributions are obtained from equation (17) by considering the interaction of a single He$^+$ ion with the laser pulse described in figure 1. While both panels relate to the same azimuthal detection angle, $\varphi_p = 0.5\pi$, they are for different polar angles: $\theta_p = 0.46\pi$ and $\theta_p = 0.5\pi$ for the upper and lower panel, respectively. In the upper panel, we observe three distinctive structures in the spectrum. The low-energy part (cyan curve), that spans the region from 0 to 60 eV, is the least pronounced and consists of fast oscillations. This is in contrast to the remaining structures. They appear as big lobes with maxima centered at either 2.96 keV in the case of the mid-energy pattern (blue curve) or 15.5 keV in the case of the high-energy one (red curve). The latter is by nearly two orders of magnitude more pronounced and, as we have checked for this particular emission direction, it contributes the most to the energy-integrated probability distribution. In the lower panel of figure 3, which is for a slightly different polar angle, the high-energy structure is missing. Thus, the structure appears to be very sensitive to the polar angle $\theta_p$. On the other hand, the low- and mid-energy patterns stay nearly the same. Such behavior can be related to radiation pressure, as we elaborate this next (for a discussion of radiation pressure, see, for instance, [65]).

Figure 4 shows the color mappings of the energy and polar angle distributions of photoelectrons from either the mid- (upper panel) or high-energy (lower panel) regions obtained for $\varphi_p = 0.5\pi$. Both distributions have the cigar-like shape, even though they are displaced from each other. In the upper panel, the maximum of the distribution is located at the point $(\theta_p, E_p - m_e c^2) = (0.483\pi, 2.92$ keV) with the peak value $\mathcal{P}(p) = 8.73 \times 10^{-6}$. In the lower panel, it is located at $(0.461\pi, 15.5$ keV) with $\mathcal{P}(p) = 14.3 \times 10^{-6}$. Once again, we conclude that photoelectrons are mostly detected with high energies. Such energies are reached by absorption of many more laser photons than intermediate energies, leading to higher radiation pressure exerted on photoelectrons. This results in a displacement of the distribution towards smaller polar angles $\theta_p$, i.e., in the direction of the driving pulse propagation [65]. Also, with absorption of a higher number of photons, the distribution becomes more spread in energy. While the width of the distribution in the upper panel is roughly 0.4 keV, in the lower panel it becomes 1 keV. On the other hand, the momentum transfer from many more photons makes the electron distribution more elongated in a certain direction. For this reason, the width of the distribution in $\theta_p$ decreases from roughly 0.02$\pi$ to 0.01$\pi$ (upper and lower panels, respectively). This explains a strong sensitivity of the high-energy structure to the polar angle $\theta_p$, observed in figure 3.

It has been shown in [40] that the saddle-point analysis of the integrals in equation (6) can give some insight into the mechanism of photoionization. Therefore, in the next section, we introduce the saddle-point analysis to interpret our numerical results.

\[ \mathcal{P}(p) = \frac{\mathcal{P}(p)}{\omega_L} \]
3.1. Saddle-point analysis

In order to perform the saddle-point analysis, the spin-resolved probability amplitude of ionization (6) is written in terms of the light-cone variables. By doing so, as before, we are able to simplify equation (6) to a single integral (see, also [40]). Namely, we obtain

$$A_{\lambda,\lambda}(p) = \frac{1}{k_0} \int_{0}^{2\pi} d\phi \, e^{iG(\phi)} [M_{\lambda,\lambda}(\phi)]_{q-Q},$$

(25)

where $\phi = k^0 x$, $M_{\lambda,\lambda}(\phi)$ is defined by equation (7), and it is calculated at the point $Q = p + (q^0 - p^0)n$. Additionally, the function $G(\phi)$ in equation (25) is given by

$$G(\phi) \equiv G(g_0, g_1, g_2, h; \phi) = \int_0^{\phi} d\phi' [g_0 + g_1 f_1(\phi') + g_2 f_2(\phi') + h (f_1^2(\phi') + f_2^2(\phi'))],$$

(26)

where

$$g_0 = \frac{\rho^0 - q^0}{k_0}, \quad h = \frac{(m_e c^2 \mu)^2}{2k \cdot p}, \quad g_1 = -m_e c^2 \mu \varepsilon_j \cdot p, \quad g_2 = k \cdot p.$$ (27)

Now, given that the functions $G(\phi)$ and $[M_{\lambda,\lambda}(\phi)]_{q-Q}$ are sufficiently regular for real $\phi$ whereas $e^{iG(\phi)}$ is a fast oscillating function as compared to the remaining parts of the integrand in (25), the standard saddle-point approximation can be applied. The saddle points are obtained by solving the equation

$$\frac{dG(\phi)}{d\phi} = 0.$$ (28)

Among them, the ones that contribute to the integral in equation (25), from now on denoted as $\phi_s$, are those with $\text{Im} \, G(\phi_s) > 0$ (or, equivalently, with $\text{Im} \, \phi_s > 0$). Keeping this in mind, the probability amplitude of ionization in the saddle-point approximation becomes

$$A_{\lambda,\lambda}(p) = \frac{1}{k_0} \sum_s e^{iG(\phi_s)} \sqrt{\frac{2\pi i}{G''(\phi_s)}} [M_{\lambda,\lambda}(\phi_s)]_{q-Q}.$$ (29)

This expression suggests that the interference-dominated structures in the energy spectra of photoelectrons should appear at regions for which two or more saddle points contribute the most to the above sum. In contrast, if just one of them dominates over a broad range of photoelectron kinetic energies (i.e., if $\text{Im} \, G(\phi) < 0$ for the saddle point represented by $\phi_s$), the corresponding values of the remaining saddle points, the interference-free structures are expected to be found.

In figure 5, we show the imaginary part of $G(\phi_s)$ as a function of the photoelectron kinetic energy for four saddle points which contribute to the sum in equation (29). The laser field parameters are the same as in figure 1 and $\phi_s = 0.46\pi$ in both panels, two polar angles have been chosen: $\theta_p = 0.46\pi$ (left panel) and $\theta_p = 0.5\pi$ (right panel). Note that, for energies smaller than 60 eV, at least two saddle points contribute significantly to the probability amplitude of ionization (29) as the corresponding $\text{Im} \, G(\phi_s)$ takes the smallest values (cyan and magenta lines). As expected, this region coincides with the interference-dominated low-energy portion of the spectra presented in figure 3. Once again, we would like to stress that this is the energy domain beyond the validity of RSFA. For this reason, the analysis above can provide only a qualitative understanding of this particular interference pattern. Fortunately, for the considered laser pulse parameters and in contrast to the high-frequency case investigated in our previous studies [40, 41], the low-energy electrons contribute marginally to the total ionization yield.

The interference-free structures are expected to appear at photoelectron kinetic energies at which an isolated saddle point contributes dominantly in equation (29). To illustrate this, we compare figures 3 and 5. For $\theta_p = 0.46\pi$, the mid- and high-energy interference-free structures peak in the energy region where $\text{Im} \, G(\phi_s)$ calculated at the dominant saddle points (blue and red curves in the left panel of figure 5) reach their minimum values. The same conclusion can be drawn for $\theta_p = 0.5\pi$, with the difference that the high-energy lobe is absent in this case. To understand this we note that $\text{Im} \, G(\phi_s)$, shown as the red and blue curves in the left panel of figure 5, have their minima which are by roughly three orders of magnitude smaller than the remaining values of $\text{Im} \, G(\phi_s)$ at the same electron energies. The situation in the right panel is changed as the minimal value of the red curve increases relative to the other values of $\text{Im} \, G(\phi_s)$. Thus, the high-energy structure disappears in the spectrum (see, figure 3). Note also that the values of $\text{Im} \, G(\phi_s)$ for the saddle point represented by the red curve seem to change rapidly with the angle $\theta_p$, thus, resulting in a sensitivity of the corresponding high-energy structure to the polar angle $\theta_p$. This is in contrast to the blue curve, which agrees with our earlier observation that the mid-energy lobe in figure 3 is less sensitive to the change of $\theta_p$.

The saddle-point analysis gives a reasonable estimate of the photoelectron time of emission, which is related to $\text{Re} \, \phi_s$ (see, e.g., [40]). Specifically, the two saddle points causing the low-energy interference pattern in figure 3 are such that, for the electron kinetic energies up to 60 eV, $\text{Re} \, \phi_s$ changes as 0.28–0.38 (cyan line) and 5.78–5.87 (magenta line). This indicates that the corresponding photoelectrons are ionized at the beginning and at the end of the driving pulse. The mid-energy interference-free structure, on the other hand,
peaks at $\Re \phi_f = 1.42$ (blue line). These electrons are emitted more towards the middle of the ramp-up part of the pulse. Finally, the high-energy structure has its maximum at $\Re \phi_f = 3.7$ (red line). Thus, the corresponding electrons are ionized at roughly maximum value of the laser pulse envelope.

Finally, let us analyze the time-dependent ponderomotive energy of an electron undergoing the quiver motion in a laser field,

$$U_p(\phi) = \frac{e^2 A^2(\phi)}{2m_e}.$$  

(30)

For the laser field defined in Figure 1, $U_p$ is plotted in Figure 6. Hence, we see that photoelectrons which contribute to the mid- and high-energy structures appear in the continuum at times when the ponderomotive energy equals 3.36 keV and 15.41 keV (intersection of the blue and red vertical lines with the curve in Figure 6), respectively. Note that those values are close to the final photoelectron kinetic energies for which the interference-free distributions acquire their maximum values, i.e., 2.92 and 15.5 keV (see the discussion below Figure 4). This suggests that, for the mid- and high-energy photoelectrons, the ponderomotive energy acquired at the moment of their birth in the continuum is basically transferred into their longitudinal motion. Such an argument cannot be made for low-energy electrons. This discrepancy is actually expected, as the RSFA offers only a qualitative description of photoionization in this part of the spectrum.

Since the RSFA is particularly suitable to describe the highly energetic ionization, in the remaining part of this paper we focus on the high-energy interference-free structure that appears in the photoelectron spectrum of ionization. As we show next, it makes a potential for generating short-in-time electron wave packets.

4. High-energy photoelectron wave packets

To generate photoelectron wave packets, $\Psi_{\lambda,\lambda}(x)$, one has to superimpose the elementary electron waves, with $A_{\lambda,\lambda}(p)$ (equation (12)) defining the profile of the wave packet. Note that the probability amplitude of ionization $A_{\lambda,\lambda}(p)$, that enters (31), takes, in general, complex values. Therefore, the global phase of $A_{\lambda,\lambda}(p)$ has to be a sufficiently regular function of $p$ to guarantee that the plane waves in (31) interfere constructively.

4.1. Global phase of the probability amplitude of ionization

The spin-resolved probability amplitude of ionization $A_{\lambda,\lambda}(p)$ can be represented as

$$A_{\lambda,\lambda}(p) = \exp[i\Phi_{\lambda,\lambda}(p)]|A_{\lambda,\lambda}(p)|,$$  

(32)

where the global phase equals $\Phi_{\lambda,\lambda}(p) = \arg[A_{\lambda,\lambda}(p)]$. In our further analysis, we will consider electron wave packets propagating in a given space direction determined by the fixed polar and azimuthal angles. For this reason, we will focus below on the energy dependence of the global phase, $\Phi_{\lambda,\lambda}(p)$, and the amplitude modulus squared, $|A_{\lambda,\lambda}(p)|^2$, proportional to $P_{\lambda,\lambda}(p)$.

As mentioned above, in order to successfully synthesize an electron wave packet, the global phase of the ionization probability amplitude has to be regular. To quantify this statement, we expand the global phase in a Taylor series around the maximum of the high-energy distribution. Hence, we obtain a constant term, which is physically irrelevant, as well as the linear and quadratic terms, which introduce the time delay and chirp of the electron wave packet, respectively. In order to eliminate the chirp, the derivative of the global phase with respect to the photoelectron energy has to be nearly constant. Furthermore, this derivative should be spin-independent in order to guarantee that the time delay of the electron wave packet does not depend on the initial and final spin degrees of freedom. Note that, in our analysis, the initial spin state is assumed to be projected on the direction of laser pulse propagation, whereas the final spin state is projected on the direction of electron propagation, i.e., it is defined as the helicity.

Figure 7 shows the derivative of the global phase $\Phi_{\lambda,\lambda}(p)$ with respect to the electron energy (upper panel) and the spin-fixed probabilities of ionization, equation (16) (lower panel), as functions of the photoelectron kinetic energy. The laser field parameters are the same as in Figure 1, whereas the polar and azimuthal detection angles are $\beta_p = 0.46\pi$ and $\varphi_p = 0.5\pi$, respectively. The four curves correspond to different initial and final spin degrees of freedom of the electron ($\lambda_i, \lambda_f$): $(-, +)$ (dashed red line), $(+, -)$ (dashed cyan line), $(-, +)$ (solid green line), and $(+, +)$ (solid blue line). In the upper panel, all curves overlap, meaning that the derivatives of all phases are spin-independent. We see that these derivatives are nearly constant over a broad range of the final electron kinetic energies. This indicates that no chirp should be observed in the synthesized electron wave packets. Note also that all spin-resolved probabilities shown in the lower panel are comparable, even though the spin-flipping processes are preferable.
contains all irrelevant constants and the electron emission angles are indicated in the upper panel. The four energy and integral only. Thus, the space dimensional integral in equation contains a range of free-electron energies. This can be done by multiplying \( \tilde{\varphi}_l \), which on the scale presented in the upper panel occur as a blue area. Since the real and imaginary parts of \( A^{(1)}(t, d = 34400\alpha_0) \) are shifted in phase by roughly \( \pi/2 \), the corresponding probability \( P^{(1)}(t, d = 34400\alpha_0) \), plotted in the upper panel as a solid yellow line, does not oscillate in time.

4.2. Space–time distributions

Keeping in mind the above discussion, we go back to equation (31). Now, if we want to analyze the electron wave packet propagating in a given space direction \( n_0 \), defined by the polar and azimuthal angles \( \theta_0 \) and \( \varphi_0 \), respectively, we have to introduce in (31) constrains on possible values of \( A_{\lambda_i}(p) \). This can be done by multiplying \( A_{\lambda_i}(p) \) by the properly chosen filter function [40],

\[
F(p) = \theta(E_{\text{max}} - E_p + me^2) \times \theta(E_p - me^2 - E_{\text{min}}) \delta^2(\Omega_p - \Omega_{n_0}). \tag{33}
\]

In doing so, we create the electron wave packet that propagates in the direction \( n_0 \) (with \( \theta_0 = \theta_p \) and \( \varphi_0 = \varphi_p \)) and contains a range of free-electron energies \( E_p \), namely, \( E_p \in [E_{\text{min}} + me^2, E_{\text{max}} + me^2] \). This reduces the three-dimensional integral in equation (31) to the one-dimensional integral only. Thus, the space–time probability amplitude of ionization (31) becomes

\[
\tilde{A}_{\lambda_i}(t, d) = \mathcal{N}_A \int_{E_{\text{min}} + me^2}^{E_{\text{max}} + me^2} dE_p e^{-i E_p t - i E_p d} |\varphi_l(p)| \tilde{\varphi}_l(\Omega_p - \Omega_{n_0}) \times \sqrt{E_p} |p| A_{\lambda_i}(p|n_0), \tag{34}
\]

where the factor \( \mathcal{N}_A \) contains all irrelevant constants and \( d = n_0 \cdot x \) is the electron distance from the parent ion. In the following, we extract from \( \tilde{A}_{\lambda_i}(t, d) \) the common phase factor \( e^{-im_x c^2 t/\hbar} \) and define

\[
A_{\lambda_i}(t, d) = e^{im_x c^2 t/\hbar} \tilde{A}_{\lambda_i}(t, d). \tag{35}
\]

This time-dependent factor artificially introduces rapid oscillations into the probability amplitude and is irrelevant when calculating probabilities.

Note that \( A_{\lambda_i}(t, d) \) is a four-component object due to the presence of the free-electron bispinor \( u^{(+)}_p \). However, in the following, we will concentrate on its first component, \( A^{(1)}(t, d) \). This is because the space–time probability distributions, defined for each component \( j = 1, \ldots, 4 \) as \( P_p^{(j)}(t, d) = |A^{(j)}(t, d)|^2 \) and with their maximum normalized to 1, are nearly identical, although the phases of \( A^{(j)}(t, d) \) are different.

In figure 8, we analyze the first component of the ionization probability amplitude (35) for the case when the initial and final electron spins are antiparallel to the laser pulse and the electron propagation directions, respectively; in our notation, \( A^{(1)}(t, d) \). The results have been obtained for a distance \( d = 34400\alpha_0 \) from the parent ion, where \( \alpha_0 \) is the Bohr radius. This particular distance has been chosen such that the electron wave packet has just left the laser pulse.
\[ \phi = k \cdot x \approx \omega t > 2\pi. \] Moreover, we have taken \( E_{\text{min}} = 7 \text{ keV} \) and \( E_{\text{max}} = 18 \text{ keV}. \) Note that \( A^{(1)}(t, d = 34400a_0) \) is a rapidly oscillating function of \( t, \) which is illustrated in the upper panel by plotting its real part. To show these fast oscillations more clearly, in the lower panel, we plot both real (solid blue) and imaginary (dashed red) parts of \( A^{(1)}(t, d = 34400a_0) \) in a very short time interval. They present either the sine or cosine type of behavior, that lead to a smooth oscillation-free behavior of the corresponding probability of ionization, \( \mathcal{P}^{(1)}(t, d = 34400a_0). \) The latter is presented in the upper panel as a solid yellow line.

Note that, even after extracting the phase related to the rest energy of the electron from the probability amplitude (see equation (35)), the latter is still a rapidly oscillating function of time. This follows from the fact that the electron wave packet is built up from the high-energy interference-free portion of the spectrum, for which the condition \( E_p - m_e c^2 \gg \omega \) is met. Furthermore, we have determined that the resulting electron pulse is very long, as it comprises at least few thousands of oscillations. It is known from laser physics that, for long pulses, the so-called carrier-envelope phase (CEP) is irrelevant. Hence, the same can be expected for the electron diffraction experiments. It is anticipated that the change of CEP for the remaining components of the probability amplitude will not play a significant role either.

Finally, we have estimated the width of the electron pulse. As it follows from the upper panel of figure 8, just after leaving the laser focus and before spreading in time, the electron pulse width is roughly \( \Delta t \approx \pi/(10\omega) \approx 400 \) as.

### 4.3. Analysis for larger intensities

Consider a more intense laser field. It is expected that, for larger intensities of the driving laser field, the resulting high-energy portion of the photoelectron spectrum is shifted towards higher energies. This follows from the fact that the time-dependent ponderomotive energy (30) increases linearly with intensity when the remaining laser field parameters are kept unchanged. Thus, for larger intensities, there is more energy transferred into the longitudinal motion of photoelectrons (see, our analysis in section 3.1). It is not obvious, however, how the maximum and the width of this distribution change with increasing the laser field intensity. For this reason, we consider now the case when the photoelectrons are released by the laser field of time-averaged intensity \( I = 10^{18} \text{ W cm}^{-2}. \) In order to check the consistency of the RSFA for this laser intensity we have estimated, using the Monte Carlo method, that the total ionization probability equals \( 3.5 \times 10^{-4} \) with the standard deviation smaller that 3.5%. This calculation was performed assuming that \( E_{\text{max}} = 200 \text{ keV} \) in equation (24).

In the upper panel of figure 9, we demonstrate the spin-resolved probability distribution of ionization (16) calculated for the spin configurations \((\lambda, \lambda); (-, -) \) (solid blue line) and \((- , +) \) (dashed red line) as a function of the photoelectron kinetic energy. While the averaged intensity of the laser pulse is \( I = 10^{18} \text{ W cm}^{-2}, \) its remaining parameters are the same as in figure 1. Moreover, the distributions are for the polar and azimuthal detection angles \( \theta_p = 0.38\pi \) and \( \varphi_p = 0.5\pi, \) respectively. As argued in section 3, the high-energy structure observed in the ionization probability distribution is very sensitive to the angle \( \theta_p. \) By comparing figures 4 and 9, we observe that in the current case the high-energy photoelectrons are detected at a much smaller polar angle than in figure 4. As the driving field is now more intense, it exerts a stronger radiation pressure on the photoelectrons. As a result, the respective probability distribution is shifted towards the direction of propagation of the laser field, i.e., towards smaller \( \theta_p. \) Moreover, by comparing \( \mathcal{P}_{\ldots}(p) \) and \( \mathcal{P}_{\pm}(p) \) for \( I = 10^{18} \text{ W cm}^{-2} \) (upper panel of figure 9) with the corresponding results for \( I = 10^{17} \text{ W cm}^{-2} \) (lower panel of figure 7), we see that the maximum of the probability distributions is now shifted by one order of magnitude towards larger photoelectron kinetic energies. The shifting is proportional to the increase of the laser pulse intensity (or the ponderomotive energy). Finally, we observe that the energy bandwidth is wider for the distributions presented in figure 9 than for those in figure 7. This indicates that the electron pulses build up from these distributions should be shorter for the former. To illustrate this, in the lower panel of figure 9, we present the corresponding space–time probability distributions of the photoelectron wave packet \( \mathcal{P}_{\ldots}(t, d) \)
(solid blue line) and \( P_{\perp}(t, d) \) (dashed red line), calculated at a distance \( d = 91000a_0 \) from the parent ion. Both distributions, defined as

\[
P_{\perp}(t, d) = |A_{\perp}(t, d)|^2 A_{\perp}(t, d),
\]

are scaled to the maximum of \( P_{\perp}(t, d = 91000a_0) \). Now, the temporal width of the photoelectron pulse is roughly \( \Delta t \approx 0.025\pi/\omega \), which is around four times shorter than for the case studied in figure 7. Note that the time duration of the electron pulse does not scale with the incident laser pulse intensity, as the position of the maximum of the probability distribution does. Actually, the ratio of the energy bandwidth to the energy position at which we observe the maximum decreases with increasing the laser intensity. In other words, the structure, if scaled to the most probable energy of emitted electrons, becomes more narrow. As a consequence, the photoelectron pulse which leaves the laser focus lasts in the current case for roughly 100 as.

Note that the above discussion was based on the plane-wave-fronted pulse approximation for the driving laser field. Next, we will elaborate on the validity of this approximation in relation to the laser pulse parameters used in this paper.

4.4. Validity of the plane-wave-fronted pulse approximation

Consider a laser pulse of power \( P_{\text{pulse}} = 10 \, \text{PW} \), which is focused such that its time-averaged intensity distribution perpendicular to the laser pulse propagation is of the Gaussian form,

\[
I(r_z) = I \exp \left( -\frac{r_z^2}{2\sigma_L^2} \right).
\]

Here, \( I \) is the laser beam peak intensity whereas \( \sigma_L \) determines the spatial size of the focus in the transverse direction. Thus, the total power of the pulse equals

\[
P_{\text{pulse}} = \int d^2r_z \, I \exp \left( -\frac{r_z^2}{2\sigma_L^2} \right) = 2\pi \sigma_L^2 I.
\]

If \( I = 10^{18} \, \text{W cm}^{-2} \), then

\[
\sigma_L^2 = \frac{P_{\text{pulse}}}{2\pi I} = \frac{10^{-2}}{2\pi} \text{cm}^2 \approx \frac{1}{(25)^2} \text{cm}^2,
\]

leading to \( \sigma_L \approx 4 \times 10^{-4} \, \text{m} \). We also estimate that for 10 TW pulses of time-averaged intensity \( I = 10^{17} \, \text{W cm}^{-2} \), the transverse size of the laser focus \( \sigma_L \) is by one order of magnitude smaller. In order to test the validity of the plane-wave-fronted pulse approximation, these values should be compared with the distance at which photoelectrons escape from the focus as measured in the perpendicular direction, \( d_{\text{escape}} \). Assuming that the position of the parent ion is not influenced by the laser field, due to the ion large mass, we estimate that \( d_{\text{escape}} \) is comparable to the separation between the electron wave packet and the ion after leaving the laser focus, as the polar angle of emission is close to \( \pi/2 \). It is also expected that \( d_{\text{escape}} \) is the largest for the most energetic photoelectrons.

Our space–time analysis presented above shows that \( d_{\text{escape}} \) does not exceed \( 10^5 a_0 \approx 5 \times 10^{-6} \, \text{m} \) for the time-averaged intensity \( I = 10^{18} \, \text{W cm}^{-2} \), and that it decreases for smaller intensities. Hence,

\[
d_{\text{escape}} \ll \sigma_L.
\]

In the current example, \( d_{\text{escape}} \) is smaller than the space-size of the laser focus by at least two orders of magnitude, provided that the total power of the pulse is around 10 PW. Note that, for 10 TW pulses and \( I = 10^{17} \, \text{W cm}^{-2} \), the space-size of the focus is \( \sigma_L \approx 4 \times 10^{-5} \, \text{m} \) and \( d_{\text{escape}} \approx 4 \times 10^4 a_0 \approx 2 \times 10^{-6} \, \text{m} \). This analysis shows that the plane-wave-fronted pulse approximation is suitable for the parameters chosen in the current paper. For lower pulse intensities, our estimations can be improved by increasing the total power of the pulse. On the other hand, for a fixed pulse power, we can extend the validity of the plane-wave-front approximation by designing the laser focus which is not cylindrically symmetric but is elongated in a particular direction. Finally, it is expected that this approximation can break for tighter focusing for which, however, the interference-free supercontinuum does not appear.

Note that the above estimations follow from the assumption that the longitudinal components of the electromagnetic field can be neglected. To justify this, we recall that the high-energy photoelectrons are ionized at the pulse maximum. After being ionized, they are marginally displaced from the symmetry axis of the Gaussian-type pulse. This means that the high-energy photoelectrons (being in the focus at all times) move close to the symmetry axis of the laser beam. Close to this axis the longitudinal components of the electromagnetic field are negligibly small and, as such, have been neglected in our analysis.

5. Geometry of ionization probability distributions in the momentum space

Above we have presented the ionization probability distributions as functions of energy, polar-, and azimuthal angles of photoelectrons. It appears, however, that another approach, which arises when studying the geometry of high-energy probability distributions of photoelectrons in the momentum space, is more convenient. This new approach can be used to select, without performing demanding numerical calculations, only those regions in the momentum space where ionization dominates. Moreover, it also clearly singles out those domains of momenta in which either interference- or interference-free patterns are observed.

5.1. Momentum spiral

As we have already noted, the dominant contribution to the ionization probability amplitude comes from saddle points \( \phi_s \)
for which the imaginary part of $G(\phi)$ is nearly zero, or which have small imaginary parts themselves as compared to the remaining saddle points. In general, to find $\phi_s$, the saddle point equation (28) has to be solved numerically. It appears, however, that for intense laser pulses an approximate solution can be found that amazingly well describes the main features of highly energetic ionization. In the following, we describe this method.

First, we choose an arbitrary real laser phase $\phi_p$, which is from the interval $[0, 2\pi]$ (at the moment we keep the subscript $p$ in order to emphasize that this phase is related to a particular photoelectron momentum but later we shall drop this subscript). Then, we define the momentum $p^\perp$, perpendicular to the laser pulse propagation direction $n$, as

$$ p^\perp = p - (p \cdot n)n = eA(\phi_p). $$

(41)

This formula allows us to relate the phase $\phi_p$ to the azimuthal angle of the electron emission,

$$ \varphi_p = \arg[eA(\phi_p) \cdot (\epsilon_1 + i\epsilon_2)], $$

(42)

where $\arg(z) \in [0, 2\pi]$ is the phase of a complex number $z$. Next, we define the parallel component of $p$ as

$$ p^\parallel = p \cdot n = \frac{(p^\perp)^2}{2q^0} = \frac{e^2A^2(\phi_p)}{2q^0}. $$

(43)

In order to satisfy the saddle-point equation (28) we have to assume that

$$ p^0 = q^0 + p^\parallel = q^0 + \frac{e^2A^2(\phi_p)}{2q^0}. $$

(44)

However, the four-momentum constructed in this way is not on the electron mass shell. One can check that

$$ p \cdot p = (p^0)^2 - p^2 = (q^0)^2 = mc^2(1 - Z^2\alpha^2), $$

(45)

which only for light atoms (i.e., when $Z^2\alpha^2 \ll 1$) is nearly on the mass shell. Therefore, for the photoelectron momentum,

$$ p(\phi_p) = p^\parallel n + p^\perp = \frac{e^2A^2(\phi_p)}{2q^0} n + eA(\phi_p), $$

(46)

for which the on-mass-shell energy is equal to $E_p = \sqrt{(mc^2)^2 + (cp)^2}$, the real phase $\phi_p$ is only an approximate solution of the saddle point equation (28).

Our numerical analysis shows that, for sufficiently intense laser pulses and light atoms, one of the solutions of equation (28) with momentum defined by (46) is very well approximated by the real phase $\phi_p$. While changing it, we observe that the tip of the momentum defined in equation (46) draws a spiral in the three-dimensional momentum space, which crucially depends on the laser pulse shape. Its projection on the polarization plane, spanned by the vectors $\epsilon_1$ and $\epsilon_2$, becomes the curve followed by the vector potential tip; with the sign of the electron charge accounted for it is presented in figure 1. It appears that, for photoelectron momenta close to this spiral, the probability distribution acquires “local maxima”, as we show in this next section.

For ionization of light atoms, which is the case discussed in this paper, the pronounced maxima in the high-energy portion of ionization spectrum are achieved at such kinetic energies which are close to the electron ponderomotive energy $U_p(\phi_p)$. This happens because there is a saddle point $\phi_s$ such that $\phi_s \approx \phi_p$ (i.e., the one with the smallest Im $\phi_s$), for which the imaginary part of $G(\phi)$ is nearly zero. To illustrate this, in figure 10 we compare the most relevant saddle point $\phi_s$ (the one for which Im $G(\phi_s)$ acquires the smallest positive value) with the real phase $\phi_p$ (that uniquely defines the momentum of emitted electron). In the top panel, we show the imaginary part of the most relevant saddle point as a function of the phase $\phi = \phi_p$. As anticipated, Im $\phi_s$ is nearly zero.
provided that the phase \( \phi \) is not close to the boundaries, or, in other words, that the temporal ponderomotive energy \( U_p(\phi) \) is sufficiently large (much larger than the ionization potential). In the middle panel, we plot the difference between the laser phase, \( \phi = \phi_p \), and the real part of the most essential saddle point, \( \text{Re} \phi_p \). Again, for phases for which the ponderomotive energy is large, we observe excellent agreement, as \( \text{Re} \phi_p \approx \phi_p \). At this point it is important to recall that, for high-energy ionization, the ponderomotive energy gives a very good estimate of the kinetic energy of photoelectrons. We conclude, therefore, that the big lobes observed in the high-energy portion of the ionization spectrum (see, section 3) are indeed defined by the real phase \( \phi_p \). Finally, in the bottom panel of figure 10 we present the imaginary part of \( G(\phi) \) for the relevant saddle point (blue line) and one of the remaining saddle points (red line) as a function of \( \phi = \phi_p \). This other saddle point has been chosen since for some \( \phi_p \) it also leads to a nearly zero value of \( \text{Im} G(\phi) \). Specifically, this happens at the self-intersections of the spiral representing the vector potential, \( \mathbf{A}(\phi) \), in figure 1. In such cases we observe the interference pattern in ionization, as discussed in section 5.2. Note that \( \text{Im} G(\phi) \) for the remaining saddle points is by roughly three orders of magnitude larger than what is shown in the bottom panel of figure 10. Hence, the contribution of the remaining saddle points to the probability amplitude \( A_{\lambda\lambda}(p) \) is negligible.

It is important to realize that the momentum spiral discussed in this section can lead to valuable predictions for the energy and polar-angle distributions of photoelectrons. For instance, by inspecting figure 4 where some of the energy-polar-angle probability distributions are presented, we can localize their maxima for the electron energies given by equation (44). Using this equation, we can also determine their corresponding polar angles,

\[
\cos \theta_p = \frac{p^0 - q^0}{|p|} = \frac{E_p - E_0}{\sqrt{E_p^2 - (m_e c^2)^2}}.
\]

As we have checked, the above equation very well reproduces the polar angles at which the maxima in the ionization probability distributions appear.

In figure 10, we have compared the most relevant saddle point \( \phi_p \) and its real approximation \( \phi_p \) for the moderate laser pulse intensity \( I = 10^{17} \text{ W cm}^{-2} \). We have also checked that, with increasing the intensity, this approximation is improved. On contrary, with decreasing the intensity, the actual saddle points start to significantly deviate from our approximate solution of the saddle-point equation. This proves that our analytical approach cannot be successfully used for smaller intensities.

5.2. Ionization probability distributions in the vicinity of the spiral

In figure 11, we present the one-dimensional probability distribution, \( P(\mathbf{p}(\phi)) \) (see, equation (46)), calculated along the momentum spiral. It does not change drastically, except for phases for which the interference pattern is observed. For our further purpose, we define the two-dimensional distribution of ionization in the momentum plane perpendicular to the curve \( \mathbf{p}(\phi) \). First, we define the two-dimensional distribution of ionization in the momenta plane perpendicular to the curve \( \mathbf{p}(\phi) \). We have also checked that, for large intensities, this distribution is focused even more tightly around its center. Meaning that, while \( p_x \) and \( p_y \) acquire larger values with increasing the driving field intensity, they increase slower than \( |\mathbf{p}(\phi)| \). This happens also for phases \( \phi \)
for which the interference pattern starts developing. We conclude therefore that, for intense laser pulses, highly energetic ionization predominantly occurs with momenta such that their tips lie within a narrow region surrounding the momentum spiral, and that the extent of this region relatively decreases with increasing the laser pulse intensity. Furthermore, outside this region the ionization probability is marginally small. This is the reason why the high-energy supercontinua appear at specific directions of electron detection, as presented, for instance, in figure 3.

6. High-energy interference patterns

Typically, the high-energy portion of the ionization probability distribution does not exhibit the interference pattern, in contrast to its low-energy counterpart. Still, even for high energies and for selected directions of photoelectron detection, the interference of probability amplitudes can be observed. It is the momentum spiral which allows one to precisely determine the domains of electron momenta for which the interference pattern occurs. These are the regions closely surrounding the self-intersections of the three-dimensional momentum spiral. This has already been demonstrated in figure 11. The analytical formula for \( p(\phi) \), given by equation (46), predicts that for \( I = 10^{17} \, \text{W cm}^{-2} \), one can expect the strongest interference for \( \theta_p = 0.46\pi \), \( \varphi_p = \pi \), and for the kinetic energy \( E_p - m_e c^2 \) close to 11 keV. This is indeed the case presented in figure 13.

6.1. Coherent double-hump electron wave packets

Interference of probability amplitudes of ionization is observed if at least two saddle points contribute predominantly to the process. For a symmetric laser pulse considered in this paper, there are two such saddle points in the high-energy portion of the photoelectron spectrum, as indicated in the bottom panel of figure 10. Hence, in the case of constructive interference, we should expect enhancement of the probability distribution by a factor of four. This is indeed the case demonstrated in figure 11. It also means that the same photoelectron can be created at two different times, which should be reflected in the space–time probability distribution. To illustrate this, in figure 14 we show the wave packet synthesized from the energy distribution plotted in figure 13. It consists of two humps, each of roughly 400 as duration, with maxima acquired at times at which ionization occurs with the largest probabilities. Since the kinetic energies, \( E_p - m_e c^2 \), are much smaller than the electron rest energy, these two structures spread rapidly in time. Just after leaving the laser focus (at the distance 30000 \( a_0 \) from the parent ion) the two humps overlap and start to interfere. This ability to interfere is a clear signature of the coherence of the single-electron double-hump wave packet generated in this process. For much larger laser pulse intensities, when the electron is emitted with higher energies, the wave packet spreading is slower. Moreover, as already discussed in [68], the application of finite train of laser pulses, sufficiently well separated from each other, can generate the single-electron coherent train of wave packets that do not overlap at large

![Figure 12](image1.png)

**Figure 12.** Color mapping of the probability distribution of ionization in the plane perpendicular to the spiral \( p(\phi) \) (see, equation (52)) for \( \phi = \pi \). Note that the distribution is tightly focused around the momentum \( p(\phi) \). This, in turn, indicates that the spin-averaged probability distribution \( \mathcal{P}(p) \) (equation (17)) is dominant for momenta closely surrounding the spiral \( p(\phi) \). The laser field parameters are the same as in figure 1.

![Figure 13](image2.png)

**Figure 13.** Ionization probability distribution \( \mathcal{P}(p) \) for \( \theta_p = 0.46\pi \), \( \varphi_p = \pi \), and \( I = 10^{17} \, \text{W cm}^{-2} \). In the upper panel, the distribution is plotted in a broad energy range. It exhibits a regular interference pattern, the details of which are shown in the lower panel. As we have checked, the peaks are nearly equidistant and separated by \( \omega_L = N_{occ} \omega \). Note, however, that this is not a typical situation for ionization by such short laser pulses \( (N_{occ} = 3) \).
distances. This means that, in principle, it would be possible to study experimentally the ‘pump-and-probe’ diffraction patterns with the same electron.

In closing let us discuss the longitudinal coherence which is a standard concept in the area of electron pulse generation. Typically, it is defined as $\ell_c = v_0/(2\Delta E)$, where $v_0$ is the longitudinal velocity and $\Delta E$ is the energy bandwidth of the pulse. This quantity determines the shortest distance over which the electron pulse is coherent [24]. We believe, however, that this definition is meaningless for wide-bandwidth pulses, as the ones considered in our paper. The reason being that this definition does not carry any information about phases of elementary waves which interfere constructively to build the pulse. It is crucial for constructive interference that the elementary waves are phase-locked (i.e., their phase derivative has to be approximately constant as a function of the electron energy). This is in analogy to generation of attosecond light pulses out of the wide-bandwidth high harmonics. As long as we talk about electron pulses of a narrow bandwidth, one can assume that the phases are nearly the same. In such case, the definition of longitudinal coherence is meaningful. For wide-bandwidth pulses, however, $\ell_c$ would be very small even if the electron waves were phase-locked. Specifically, for the data presented in our paper, this definition gives $\ell_c$ of the order of $a_0$. We suspect, however, that the electron waves add constructively over much larger distances that it follows from the uncertainty principle.

We would like to stress that in our case the electron states are phase-locked. This may lead to a time delay of the synthesized electron wave packets but does not influence their coherent properties on distances much larger than the longitudinal coherence length estimated from the uncertainty relation. Of course, like in other theoretical studies, we consider an ideal physical situation. For this reason, the coherent properties of electron wave packets are preserved even at very large distances. However, in real experimental situations electrons interact, for instance, with the laser-generated plasma in a rather uncontrolled way. Due to this and other circumstances the electron phase properties will be disturbed. Our investigations show that if such a distortion is maximal, i.e., the phase changes randomly between $-\pi$ and $\pi$ then, instead of the regular double-hump structure presented in figure 14, we observe a randomly distributed signal in which all coherent properties of the pulse are washed out. On the other hand, if a small random component is added to the actual phase, the coarse-grained double-hump structure will survive. Such analysis is, however, beyond the scope of this paper and is going to be presented elsewhere.

7. Conclusions

Using the relativistic framework of strong-field approximation developed in [40], we have studied ionization of He$^+$ ions by intense laser pulses. The latter have been treated in the plane-wave front approximation. We have shown that this approximation is suitable to describe ionization by not tightly focused pulses, like the ones considered in this paper.

In addition to our earlier works [40, 41], we have demonstrated a possibility of generating multiple supercontinua in the energy spectra of photoelectrons. This is possible using relativistically intense laser pulses of circular polarization. Such intense pulses result in producing very energetic photoelectrons, whose final kinetic energy is practically determined by the ponderomotive energy that they acquire from the laser field at the moment of ionization. Specifically, for the parameters used in this paper, we have shown that two supercontinua spanning the electron kinetic energies of keV and tens of keV are produced. As it has turned out, the high-energy supercontinua are created in very small polar-angular windows. Moreover, they can be shifted towards even larger kinetic energies if one applies a more intense driving pulse. In this case, also their bandwidth increase. As we have shown, this high-energy broad structures can lead to synthesis of attosecond electron pulses.

In general, photoelectron energy distributions crucially depend on the incident laser pulse parameters such as the frequency, number of cycles, polarization properties, etc. This is also clear when comparing the results presented in [40, 41] and in the current paper. In [40, 41], we have studied ionization by the high-frequency laser pulse and we have observed, for instance, that the resulting high-energy lobes in the photoelectron energy spectrum are comparable in magnitude.

Figure 14. Normalized space–time probability distribution synthesized from the energy spectrum presented in figure 13 for two distances from the parent ion, as depicted in each panel. While for a smaller distance (upper panel) two well separated lobes are observed, for a larger distance (lower panel) we detect the interference structure between the lobes; the latter is due to the spreading and coherent properties of the generated single-electron wave packet.
with the low-energy structures. On contrary, for a small-frequency laser pulse considered in this paper, the corresponding low-energy structures are dramatically suppressed. When it comes to the position of supercontinua, they are determined by the temporal ponderomotive energy. As we show in this paper, their positions scale linearly with the temporal ponderomotive energy. If we fix the averaged intensity of the laser pulse, the ponderomotive energy will be larger for smaller pulse frequencies, pushing the supercontinua towards higher photoelectron energies. Therefore, one can expect that the most favorable conditions for the generation of high-energy supercontinuum and, hence, attosecond electron pulses are met for low-frequency laser fields.

We have studied the geometry of momentum distributions of photoelectrons generated in high-field ionization. As it follows from our analysis, it is determined by the momentum spiral, \( p(\phi) \), which argument \( \phi \) corresponds to the real part of the most essential saddle point \( \phi_0 \); the latter being determined such that the imaginary part of \( G(\phi) \) is nearly zero. Along this curve, the probability amplitude acquires its maximum values, resulting in appearance of supercontinua. As we have also shown, the photoelectrons are mostly emitted with momenta which closely surround the spiral. Additionally, with increasing the intensity (and, consequently, the energy of electrons), the respective region around the spiral becomes increasingly narrower relative to \(|p(\phi)|\). This finding allows us to determine precisely positions of supercontinua both in energy and angular domains. Moreover, it singles out momenta for which the interference pattern is expected to appear. Although the interference of probability amplitudes occurs rather occasionally, it leads to the coherent single-electron wave packets consisting of two ultrashort humps. This opens the possibility of studying the ‘pump-and-probe’ diffraction patterns with the same electron.

As in all similar theoretical analyses, we have assumed here the infinite contrast of the laser pulse. Since the laser pulse may have a weak pre-pulse, which also contributes to the overall ionization signal, the question arises: what real contrast would allow the electrons to survive in the \( \text{He}^+ \) ions until the high field arrives? To answer this question, let us refer to experimental results on the double ionization of \( \text{He} \) that were published in [69, 70]. This experiment shows that for much longer pulse (roughly 100 longer pulse duration) of a similar wavelength (780 nm), and for the intensity \( 10^{15} \ \text{W cm}^{-2} \), the ratio of \( \text{He}^{2+} \) to \( \text{He}^+ \) ions is about \( 10^{-3} \). This suggests, in fact, that most of the \( \text{He}^+ \) ions would survive such a pre-pulse. Based on this experiment, one can judge that a contrast of \( 10^{-2} \) would, in principle, allow the electrons survive in \( \text{He}^+ \) ions. Note that this estimate can be too pessimistic as the ratio \( \text{He}^{2+}/\text{He}^+ \) seems to saturate for intensities larger than \( 10^{15} \ \text{W cm}^{-2} \) (see, figure 3 of [69]). Hence, we expect that the contrast would be even lower (lets say \( 10^{-1} \)) in order to corroborate our results. (Note that the theoretical analysis presented in figure 6 in [70] predicts that the aforementioned ratio of the \( \text{He} \) ions is by two orders of magnitude smaller than the experimental one.)

Similar to our previous studies [40, 41], our numerical calculations have been performed for \( \text{He}^+ \) ion, i.e., the lightest hydrogen-like ion one could possibly choose. This is because for light targets we have found a very appealing interpretation of our numerical results, that we have presented in section 5.1. One can expect that for heavier ions, the total ionization probability (with all electrons stripped off) is much smaller than one. Hence, one can speculate that the optimal conditions for generation of the high-energy photoelectrons significantly depend not only on the laser field parameters, but also on the atomic number \( Z \) characterizing the target. Thus, in order to provide a real directive role for experiments on attosecond pulse generation it can be of interest to perform a systematic analysis of how the electron pulse generation depends on the target. This problem is going to be discussed elsewhere.

In closing we note that, if ionization is driven by a finite train of laser pulses, one can expect to generate a finite sequence of electron pulses, similarly to the coherent combs investigated for the Thomson and Compton [71, 72], and for the Breit–Wheeler processes [73]. Let us also mention that interference effects related to ionization assisted by two (or more) laser pulses of different shapes and arbitrarily delayed with respect to each other (similar to the ones studied in [74] for the Breit–Wheeler process) can be also explored in the present context. Thus, ionization by relativistically intense laser pulses or by their trains can be used to engineer coherent electron pulses that are arbitrarily delayed and have different intensities. As the central energy of electron wave packets scales linearly with the time-averaged laser intensity, the generation of coherent electron beams in the MeV region can, in principle, be achieved in ELI and XCELS [42, 43]. In this context, the Compton effect studied theoretically in [75] can be also investigated experimentally. Moreover, the sensitivity of the high-energy structures to the laser pulse parameters and to the ejection direction of photoelectrons can be used in the experimental diagnosis of extremely intense and short laser pulses, in addition to other methods exploring ionization spectra [68, 76]. Furthermore, with increasing the laser pulse duration (i.e., for larger \( N_{\text{osc}} \), the rings of the momentum spiral approach each other and the interference patterns is expected to be established, at least partially. This can be avoided either by increasing the laser intensity or by applying the elliptically polarized pulses. These topics have been discussed in [77, 78].

Acknowledgments

This work is supported by the National Science Centre (Poland) under Grant No. 2014/15/B/ST2/02203. We thank Antonino Di Piazza for drawing our attention to the attoelectron diffraction physics and to Martin Centurion for fruitful discussions about ultrashort electron pulse generation.
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