Probing the influence of the Coulomb field on atomic ionization by sculpted two-color laser fields

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New Journal of Physics 15 (2013) 043050 (19pp)
Received 18 December 2012
Published 30 April 2013
Online at http://www.njp.org/
doi:10.1088/1367-2630/15/4/043050

Abstract. Interpretation of electron or photon spectra obtained with strong laser pulses that may carry attosecond dynamical and Ångström structural information about atoms or molecules usually relies on variants of the strong-field approximation (SFA) within which the influence of the Coulomb potential on the electron trajectory is neglected. We employ two-color sculpted laser fields to experimentally tune and probe the influence of the Coulomb field on the strong-field-driven wavepacket as observed by two-dimensional electron and ion momentum spectra. By comparison of measured spectra with predictions of the three-dimensional time-dependent Schrödinger equation as well as the quasi-classical limit of the SFA, the strong-field classical trajectory model, we are able to trace back the influence of the Coulomb field to the timing of the wavepacket release with sub-cycle precision.
1. Introduction

Ultrashort intense laser pulses are a unique tool to create coherent electron wavepackets with sub-cycle duration via the strongly nonlinear process of tunnel ionization of atoms or molecules [1, 2]. After their creation the wavepackets are driven by the combined forces of the laser electric field and the ionic Coulomb field [3]. It is thus possible to actively control their motion in time [4, 5] and in space [6–8] by the laser electric field. When the wavepackets are driven back to the ion core, they coherently probe the parent ion on the attosecond and Ångström scale by recording suitable probe signals such as electron momenta or energies, e.g. [9–12], or spectra of photons emitted via the process of high-harmonic generation, e.g. [13–17].

Although the motion of the wavepacket after tunnel ionization is governed by the combined forces of the laser electric field and the ionic Coulomb field, experimental electron and photon spectra could be successfully explained in many cases by the strong-field approximation (SFA) [18–20] and its semiclassical variant, the ‘simple man’s model’ (SMM) [21], in both of which the influence of the ionic field on the receding or recolliding wavepacket is neglected. The importance of the Coulomb potential is by now well appreciated and has been demonstrated in many numerical simulations, e.g. [22–26]. Inclusion of the Coulomb force into the theoretical description of e.g. electron momentum spectra is not straightforward though [27]. Likewise, clearly separating and identifying Coulomb contributions in the experimental electron or photon spectra has remained a challenge.

Here, we show that a sculpted two-color laser field that allows sub-cycle tuning of the instantaneous electric field $F(t)$ provides a tool to control and modify the relative importance of the Coulomb field on the electron wavepacket. We identify Coulomb field effects in measured three-dimensional (3D) electron–ion momentum spectra generated from helium, neon and argon atoms by two-color sculpted laser fields. We gain access to the Coulomb contributions by comparing measured spectra of the longitudinal and orthogonal momentum distributions obtained for different pulse parameters, i.e. for different attosecond evolutions of the field-induced force, with predictions of the full 3D time-dependent Schrödinger equation (TDSE) and a quasi-classical trajectory model. The latter allows us to relate features of the momentum distributions with classical trajectories in the presence (or absence) of the Coulomb force. We are, thus, able to trace back the influence of the Coulomb field to the timing of the wavepacket release within the laser field cycle.
2. Experiment

We generated cycle-sculpted laser fields by coherently superimposing a strong \( \approx 30 \text{ fs} \) (full-width at half-maximum (FWHM)) 790 nm laser pulse, frequency \( \omega \), and its second-harmonic pulse, frequency \( 2\omega \), in a collinear geometry. The second-harmonic pulse was generated in a type-I BBO crystal of thickness 500 \( \mu \text{m} \). The duration of the fundamental pulse was measured by second-harmonic FROG and the duration of the \( 2\omega \) pulse was determined from cross-correlation measurements using the \( 3\omega \) signal to be approximately twice as long as the fundamental. For the measurements of both pulse durations, the propagation distance in air and the amount of glass passed before the vacuum chamber have been taken into account. The pulse energies of the two pulses were adjusted by varying the angle of reflection off a glass plate utilizing the different reflection coefficients for p- and s-polarized light such that in the focus of the two beams the field strengths were equal, taking into account the different pulse durations and the slightly different beam diameters and tighter focusing of the \( 2\omega \) pulse. The polarization directions of the two pulses were rotated to parallel by a \( \lambda/2 \) plate for 790 nm. The pulse repetition rate was 5 kHz. The temporal overlap of the two pulses was adjusted by observing the \( 3\omega \) cross-correlation signal and compensating for the different group velocities of the two colors with calcite plates and a thin pair of fused silica wedges. The relative phase of the two pulses was varied by introducing one of the wedges into the combined red and blue laser beam. This could be done with a precision of 0.1 \( \mu \text{m} \), resulting in a precision for controlling the relative phase delay of roughly 0.3 attoseconds. As both the red and blue beams were propagating collinearly, residual phase jitter is mainly introduced by beam pointing instabilities. We estimate the overall stability of the relative phase delay during the experiments to be on the few attosecond level.

In the interaction region, the total electric field of the two-color pulse can be written as

\[
F(t) = F_0 \left[ f_1(t) \cos(\omega t) + f_2(t) \cos(2\omega t + \varphi) \right].
\]  

(1)

Here, \( f_i(t), i = 1, 2 \), are Gaussian pulse envelopes normalized to a maximum value of 1, \( \varphi \) the relative phase and \( F_0 \) the peak electric field of the two colors, which is related to the pulse peak intensity as \( F_0 = \sqrt{I} \) (here and throughout the paper atomic units are used unless otherwise stated). Similar two-color fields in this \( \omega - 2\omega \) configuration have been experimentally applied to the investigation of above-threshold ionization (ATI) (see e.g. [28–30] and references therein), to control ionization and fragmentation [31–33] as well as orientation [34] of molecules, and to control interference fringes in electron momentum spectra [35]. The sub-cycle field shape can be controlled by \( \varphi \). This allows, among other possibilities, mimicking features of the field \( F(t) \) of a near single-cycle pulse while exploiting the advantage of a relatively long (\( \approx 10 \) cycle) pulse (equation (1)). For example, the maximum cycle unidirectionality, achieved for \( \varphi = 0 \), closely resembles the limit of a true single-cycle pulse of one color (‘cosine-like pulse’) with a controlled carrier-envelope phase, which for 790 nm is only achieved for pulse durations close to 2.6 fs. Varying the relative phase \( \varphi \) of the two colors allows one to sculpt the ionizing field and hence to control the emission times and motion of the emitted wavepackets on the attosecond timescale (see figure 1).

We use cold target recoil ion momentum spectroscopy (COLTRIMS) [36] to measure the 3D momentum vector of electrons and ions emerging from the interaction of a single atom with the sculpted laser field. Our detection apparatus consists of a two-stage arrangement to provide an internally cold ultrasonic gas jet of atoms, and an ultra-high vacuum chamber (\( \approx 10^{-10} \) mbar). The laser beam is focused within the vacuum chamber into the gas jet using a spherical mirror.
Figure 1. Ionization and wavepacket motion with sculpted two-color laser pulses. Electron wavepackets are emitted at the crests of the laser electric field $F(t)$. Without the influence of the Coulomb potential, an electron born at some time $t_b$ within the pulse reaches a final momentum given by the negative vector potential at birth time, $p = -A(t_b)$. By varying $\varphi$ the timing of the wavepacket’s creation as well as its motion after ionization can be controlled. (a) The shape of one field cycle for $\varphi = 0$ is unidirectional with one strong peak per laser cycle. Because the vector potential sweeps through zero during wavepacket emission, the electron spectrum is expected to be centered around zero in the absence of the Coulomb potential. The Coulomb field influences the motion of the wavepacket and leads to a distortion of the spectral shape and a shift of the spectral mean value $\langle p \parallel \rangle$. (b) For $\varphi = \pi/2$ two wavepackets per cycle are emitted at times when $A(t)$ is positive. Therefore, without the Coulomb potential the spectral mean value is shifted towards negative values. The Coulomb force leads to a distortion of the spectral shape and a shift.
with a focal length of 60 mm. The ions and electrons created during single ionization are guided by weak magnetic and electric fields to two RoentDek DLD 80 detectors situated at opposite ends of the vacuum chamber. The homogeneous electric field of 2.5 V cm\(^{-1}\) along the \(z\)-direction is produced by equidistant copper rings; the weak homogeneous magnetic field of 6.4 G (also along the \(z\)-direction) is produced by three copper coils. With these directions of the fields, the ions are accelerated over a distance of 45 cm before they reach the upper detector, and the electrons are accelerated over 5.7 cm before reaching the lower detector. We estimate the momentum resolution of our experiment to be \(\approx 0.05\) au along the \(z\)-direction for electrons and ions and \(\approx 0.05\) au along the perpendicular \(x\)- or \(y\)-direction for electrons.

Typical experimental and simulated two-dimensional (2D) momentum spectra (see figure 2) for helium feature intricate fine-scale patterns that result from both sub-cycle and intercycle interferences [35]. On a larger scale, i.e. upon averaging over the fine-scale oscillations, the spectra along the momentum directions parallel and perpendicular to the laser polarization direction (\(z\)), \(p_\parallel\) and \(p_\perp\), feature pronounced asymmetric structures. We focus on these large-scale features in the following. We will show that they provide detailed insight into the interplay between the Coulomb and the laser fields for ionizing trajectories.

Examples for the \(p_\parallel\) distribution as a function of \(\varphi\) for He and Ar are shown in figure 3. We characterize these distributions in the following in terms of their first, \(\langle p_\parallel \rangle_\varphi\), and second moments, \(\langle (\Delta p_\parallel)^2 \rangle_\varphi\) \((i = ||, \perp)\), or, equivalently, their spectral width \(\sigma_i = ((\Delta p_\parallel)^2)_\varphi^{1/2}\). The mean value of the measured spectra, \(\langle p_\parallel \rangle\), strongly varies with \(\varphi\) and shows for helium extrema for \(\varphi \approx (n + 0.3)\pi\), \(n \in \mathbb{Z}\). The pulse peak intensity for each of the two colors was \(1 \times 10^{14}\) W cm\(^{-2}\) for helium and neon, and \(2 \times 10^{13}\) W cm\(^{-2}\) for argon. The intensity for the experiments on helium was calibrated using two independent methods that both led to very similar intensity values. Firstly, we compared the positions of the sub-cycle interference fringes and ATI peaks in the ion momentum spectrum of helium, which sensitively depend on the laser intensity, to spectra calculated by solving the TDSE [35]. We estimate the precision of this calibration to be approximately 5%. Secondly, we cross-checked the obtained value by calculating the peak intensities of the red and blue beam from the measured focal spot size, pulse duration and pulse energy. The focal spot sizes were measured outside the vacuum chamber by imaging them, separated by a dichroic beam-splitter, with a CCD camera. The energies of the red and blue pulses were also measured by separating the combined beam with a dichroic beam-splitter. The reflection off the input window into the vacuum chamber was taken into account for these measurements. The intensity used for argon was determined by scaling the precisely determined intensity of the experiments on helium with the measured pulse energies and focal spot sizes used in the experiments on argon.

The relative phase \(\varphi\) was calibrated by aligning the maxima of the experimentally observed pronounced variation of the He\(^+\) yield with \(\varphi\) (see figure 3(e)), with the maxima of the yield modulation at \(\varphi = n\pi\), \(n \in \mathbb{Z}\) predicted by tunneling theory [39]. The reason for the modulation of the yield is that a pulse with \(\varphi = 0\) exhibits larger maxima of the laser electric field than a pulse with \(\varphi = 0.5\pi\) (see figure 1). This translates into a much higher ionization rate and yield per cycle for \(\varphi = 0\). A comparison of the predictions of tunneling theory with solutions of the TDSE shows a slight offset in \(\varphi\) of roughly 0.1\(\pi\) (figure 3(e)). Thus, we estimate the precision of our calibration to be on that order. Figure 3(e) additionally shows the measured modulation of the Ne\(^+\) yield, which exhibits a smaller modulation depth than the one of He\(^+\). The latter is in good agreement with the TDSE results. Note that the phase calibration based on the modulation of the He\(^+\) yield is free from errors of any spectral distortions and Coulomb influences.

New Journal of Physics 15 (2013) 043050 (http://www.njp.org/)
Figure 2. Experimental raw (a) and (b), experimental corrected (c) and (d) and simulated (e) and (f) 2D electron momentum spectrum for helium along, $p_{∥}$, and perpendicular, $p_{⊥}$, to the polarization axis ionized by a two-color pulse with relative phase $\varphi = 0$ (left column) and $\varphi = \pi/2$ (right column). All data have been integrated over $p_{y}$. The gray bars in (a)–(d) blank out regions where our detector has no resolution for electrons. The simulation employs the TDSE in a single active electron (SAE) approximation using a model potential of helium [37]. Oscillatory structures result from sub-cycle and intercycle interferences [35], emphasized here by the logarithmic intensity scale. For the experimental corrected spectra (c) and (d) a 2D Gaussian fit with its amplitude multiplied by 0.6 is subtracted from the corresponding raw spectra (a) and (b), which emphasizes the interference structures contained in the raw spectra. In this work only raw spectra as shown in (a) and (b) are analyzed.

3. Modeling the field-driven wavepacket motion

We analyze the $\varphi$-dependence of the spectra by performing both TDSE simulations as well as quasi-classical simulations. The latter allow us to pinpoint in detail the influence of the laser
Figure 3. Momentum component parallel to the laser polarization direction, $p_\parallel$, of ions created during single ionization of He (a) and Ar (b), as well as (c) the momentum mean value of helium ions and (d) the spectral width $\sigma_\parallel$ (FWHM) of He, Ne and Ar as a function of the relative phase, $\varphi$, between the $\omega$ and $2\omega$ components of the two-color laser field. For all longitudinal momentum distributions, the transverse momentum directions, $p_x$ and $p_y$, have been integrated over. Due to the negligible momentum transfer by the photon field, the momentum of the ion is the mirror image of the electron momentum [38]. In panel (c), momentum mean values from simulations using SFCT (full black) and TDSE (dotted purple) are compared with the experimental value (dashed red). (e) Yield of $\text{He}^+$ (full blue) and $\text{Ne}^+$ (dashed red) in comparison with predictions for $\text{He}^+$ of tunneling theory [39] (full gray) and the TDSE (dotted purple).

and the Coulomb fields. In line with the SFA, we neglect in the quasi-classical simulations the influence of the ionic Coulomb field on the classical trajectories following tunnel ionization. We refer to this model as the strong-field classical trajectory (SFCT) model. It corresponds to

New Journal of Physics 15 (2013) 043050 (http://www.njp.org/)
the quasi-classical limit of the SMM [21] in which path interferences are neglected. Fine-scale modulations visible in figure 3 will therefore not be reproduced. Large-scale variations of the expectation values \( \langle p_n^\parallel \rangle \) due to classical laser field effects are, however, accounted for.

Within the SFCT model the momentum spectrum observed along the polarization direction after the laser pulse has faded is given by integration of the relation \( p_\parallel = -A(t_b) \), with \( A(t) = -\int_{t_b}^{t_a} F(t') \, dt' \) the vector potential. The spectrum represents the sum over all possible birth times \( t_b \) at which the wavepacket is emitted at the origin with a probability determined by the ionization rate [39]. The birth time to momentum mapping built into the SFCT model is visualized in figure 1 for the two-color field with relative phase \( \varphi = 0 \) (figure 1(a)) and \( \varphi = \pi/2 \) (figure 1(b)). While the ionization probability, i.e. the birth time, is controlled by the instantaneous field \( F(t) \), the asymptotic momentum of the outgoing electron is given by the time integral \( A(t) \).

For \( \varphi = 0 \) the electric field \( F(t) \) is strongly forward–backward asymmetric (referred to as approximately unidirectional in the following) with one strong positive peak per laser cycle. Due to the strongly nonlinear ionization rate, wavepacket emission thus takes place mostly around the peaks of \( F(t) \) (gray area in figure 1). The vector potential \( A(t) \) is anti-symmetric, \( A(-t) = -A(t) \), and passes through zero around these times. Therefore, within the SFCT model (as in the SFA), the electron momentum distribution is predicted to be symmetrically centered around zero. For \( \varphi = \pi/2 \), the electric field resembles locally a sine shape. Wavepacket emission takes place twice per laser cycle around the extrema of \( F(t) \). In turn, \( A(t) \) features a strong forward–backward asymmetry resulting in a pronounced (negative) offset of the momentum distribution (figure 1).

4. Coulomb field influence on electron momentum distributions

The momentum distributions along \( p_\parallel \) as a function of \( \varphi \) predicted by the SFCT model, shown in figure 4(a), display strong modulations with extrema for \( \varphi = (n + 1/2)\pi \) and zero mean value for \( \varphi = n\pi, \, n \in \mathbb{Z} \). In addition to the variation of the spectral mean value with \( \varphi \), figure 4(a) shows a similar modulation of the spectral intensity (encoded in color) as the experimentally observed spectra. This is because the crests of the electric field \( F(t) \) are smaller for \( \varphi = \pi/2 \) than for \( \varphi = 0 \). Since the ionization rate is strongly nonlinear in \( F(t) \), the ionization yield is larger for a pulse with \( \varphi = 0 \) than for \( \varphi = \pi/2 \).

We show the spectral mean value \( \langle p_\parallel \rangle \) of the calculated spectra in figure 4(a) as a function of \( \varphi \) by the full black line in figure 3(c). While the experimentally observed strong modulation with \( \varphi \) is in accordance with the SFCT model, the values of \( \varphi \) for which the measured maximum and minimum mean momenta are observed do not coincide with that of the SFCT (and the SFA) prediction. A phase shift between the measured and SFCT maxima of about 0.2\( \pi \) exists (figure 3(c)), which results in a large spectral offset for \( \varphi = 0 \), for which the SFCT model predicts zero momentum. We note here parenthetically that the frequently used calibration of the phase of the field cycle in terms of the maxima of the spectral asymmetry (e.g. in [40]) neglects this phase shift and is, thus, in general not applicable. This phase shift is due to the influence of the Coulomb field. The phase shift of 0.2\( \pi \) is reasonably well reproduced by the 3D TDSE calculation for a single active electron (SAE) in a model potential of helium [37], although the maximum values of \( \langle p_\parallel \rangle \) are overestimated by the TDSE. We believe the reason for this discrepancy is mainly the intensity smearing within the laser focus present in the experiment but not in the TDSE simulations. This smearing also contributes to the suppression of interference.
Figure 4. Momentum component parallel to the laser polarization direction, $p_\parallel$, of electrons emitted during single ionization of He simulated by the SFCT model (a) and solving the TDSE (b) as a function of the relative phase, $\varphi$, between the $\omega$ and $2\omega$ components of the two-color laser field with the same parameters as in the experiment. The electron distribution is mirrored ($p_\parallel \rightarrow -p_\parallel$) for better comparison with the ion momentum spectra in figure 3(a). The spectra in (b) have been smoothed by convolving them with a Gaussian function with an FWHM of 0.8 au (applied exclusively along the $p_\parallel$-axis but not along the $\varphi$-axis) accounting for the resolution and intensity averaging, for better comparison with the data in (a) and with the experimental spectra in figure 3(a).

structures present in the simulated spectra (figures 5(a) and (b)), which may also be a source of discrepancy in the $\langle p_\parallel \rangle$ value obtained from the experiment and simulation, respectively.

Another quantitative comparison can be made for the second moment of the longitudinal momentum distribution. Its width, $\sigma_\parallel$, as a function of $\varphi$ has been extracted from the measured data for He, Ne and Ar and is shown in figure 3(d). The data for Ne have been recorded with the same pulse as the one for He. The spectral width shows maxima for $\varphi = \pi/2$ and minima for $\varphi = 0$. This finding is, at first glance, surprising as the strongest field $F(t)$ appears for $\varphi = 0$ (see figure 1) which, according to tunneling theory [41], should give rise to the broadest momentum distribution. The origin of this apparent contradiction is the strong deviation of the asymptotic momentum distribution from a Gaussian distribution assumed at the tunnel exit, as we will show in the following. The measured asymptotic longitudinal spectra for $\varphi = 0$ and $\pi/2$ for
Figure 5. Measured ion momentum spectra for He (c) and (d), and Ar (e) and (f), as well as electron momentum spectra simulated by solving the TDSE (a) and (b), mirrored about $p_\parallel = 0$ to enable comparison with the ion spectra for He, in the direction parallel to the laser polarization direction for relative phases $\varphi = 0$ (left column) and $\varphi = \pi/2$ (right column). SFCT model predictions are shown by thin dark-gray lines in all frames. In (a) and (b) smoothed numerical spectra (shown by blue lines) accounting for the resolution and intensity averaging have been overlaid on the raw numerical spectra (red lines). The uncertainty of the experimental spectra in (c)–(f) due to count statistics is represented by the width of the lines. The arrow in (f) indicates a dip in the spectrum (see text).

Both helium and argon shown in figure 5 feature additional structures, in particular a dip in the argon spectrum for $\varphi = \pi/2$ (indicated by an arrow). None of these features are reproduced by the SFCT model (dark-gray lines). Obviously, it is thus not the laser field but the simultaneous presence of the Coulomb potential that causes the spectral distortions and the variation of the shape and width of the spectra with $\varphi$.

By a comparison of the experimental distributions with solutions of the TDSE and the SFCT classical trajectory result, the origin of some of the features present in the $p_\parallel$ distributions (figure 5) can be identified. We first focus on the case $\varphi = 0$ (left column in figure 5). Due to the strong unidirectional peak field, only one wavepacket is launched per cycle (figure 1(a)). Thus, in the absence of the Coulomb potential, the momentum spectrum should be centered around zero (see the SFCT distributions shown by the gray lines in figure 5). In the experiment, we observe a considerable shift of the spectrum to positive ion momenta for helium and argon (figures 5(c) and (e)) but also for neon (not shown). This shift is a clear signature of the distortion of the outgoing and recolliding wavepackets by the Coulomb field. We note that a
similar Coulomb-induced shift has been predicted for carrier-envelope phase-stabilized few-cycle pulses with a spectrum centered around 800 nm [24, 25].

To study the influence of the Coulomb field using a classical simulation would suggest employing the classical trajectory Monte-Carlo (CTMC) method [42, 43]. Application of the CTMC to the present case, however, faces a major difficulty: the quiver radius and the distance of the tunnel exit from the ionic core are of comparable magnitude. Consequently, the electron returns in the presence of the multi-cycle pulse several times close to the ionic core with near-zero velocity. The near coalescence of the inner turning point, where the de Broglie wavelength diverges, with the region of the Coulomb singularity renders the CTMC highly sensitive to the details of ionic core potentials or restrictions in available phase space (one-dimensional versus three-dimensional models), and, as a consequence, CTMC becomes inapplicable. We note that the SFCT remains unaffected by this difficulty as the influence of the ionic core is neglected from the outset. The fact that parts of the wavepacket return to the ion core with a very low energy and thus experience a very strong influence of the Coulomb field explains the non-zero momentum value $p_{\parallel}$ for $\varphi = 0$ observed in our experiments.

We now turn to the case $\varphi = \pi/2$ (right column in figure 5), for which the electric field-cycle consists of two equally strong main peaks each of which causes the emission of a wavepacket (see figure 1). One wavepacket is emitted during the negative, and one during the positive, half-cycle. Because for both emission events the vector potential sweeps through the same range of positive values (figure 1(b)), momentum spectra within the SFCT model should be identical for both wavepackets and centered at the same positive value (gray lines in the right column of figure 5). With the Coulomb field included, however, the two wavepackets will experience different driving forces. In the present case of helium at the intensity of $1 \times 10^{14}$ W cm$^{-2}$ for each of the two colors (resulting in a peak intensity of $3 \times 10^{14}$ W cm$^{-2}$ for the combined fields), the influence of the Coulomb field is moderate and results in a broadening of the overall $p_{\parallel}$ distribution, qualitatively reproduced by the TDSE simulation (cf figures 5(b) and (d)). This broadening explains the experimentally observed counter-intuitive larger spectral width at $\varphi = \pi/2$ (figure 3(c)) described above. For argon (figures 5(e) and (f)) a lower laser intensity was used (combined peak intensity $0.6 \times 10^{14}$ W cm$^{-2}$). Accordingly, the relative strength of the field of the ionic core compared to the laser field is enhanced. We therefore surmise that the dip in the experimental spectrum (figure 5(f)) may be caused by a severe asymmetric ion-core-induced distortion of the two wavepackets born during successive half-cycles of the laser field. As a consequence, their mean momentum values, compared to their width, are shifted much further apart than for helium. This, in turn, may lead not only to a broadening of the overall spectrum, but in addition to the double-peak structure observed in the experiment.

5. Gating of the influence of the Coulomb field

We now turn to discuss expectation values characterizing the transverse momentum distributions along $p_{\perp}$. For non-zero $p_{\perp}$, a wavepacket will drift laterally during its motion within the Coulomb long-range potential on its way toward the detector. The lateral drift can be significant even on an atomic scale such that wavepackets that are driven back to the ion core will miss the center of the binding potential and pass the ion at some distance that increases with $p_{\perp}$ [44, 45].
We explore the dependence of two expectation values on $p_{\perp}$: the first one is the forward–backward asymmetry

$$A(p_{\perp}) = \frac{P_{+} - P_{-}}{P_{+} + P_{-}},$$

(2)

with $P_{\pm}$ the total number of electrons with positive and negative momentum value $p_{\parallel}$, respectively, within a slice of a given transverse momentum $p_{\perp}$. Equation (2) provides a direct signature for the deviation of the electron trajectories from those of a free electron subject to the laser field only. Accordingly, the SFCT simulation predicts $A$ to be independent of $p_{\perp}$ with maxima at $\varphi = (n + 1/2)\pi$ and zero values for $\varphi = n\pi, n \in \mathbb{Z}$. Therefore, a TDSE simulation employing a short-ranged potential without the long-range Coulomb tail, defined as

$$V_l(r) = \begin{cases} 
\frac{1}{r}, & r < r_1, \\
\frac{1}{r} \cos^2 \left(2\pi \frac{r - r_1}{r_2 - r_1}\right), & r_1 < r < r_2, \\
0, & \text{else}
\end{cases}$$

(3)

with $r_1 = 2.5$ and $r_2 = 4$ for quantum number $l = 0$, and $V_l(r) = 0$ for $l > 0$, features only a weak dependence of $A$ on $p_{\perp}$ resulting in near-vertical iso-asymmetry-lines (figure 6(a)). By contrast, the experimental data (figure 6(c)) display a pronounced $A(p_{\perp})$ dependence with tilted isolines, i.e. a negative slope $dp_{\perp}/d\varphi < 0$ of the contour lines, in particular for $p_{\perp} < 0.2$ au (indicated by the gray dashed line in figure 6(c)). The TDSE simulation employing a model potential featuring both the long-range Coulomb tail as well as the short-ranged modifications of the He$^+$ ion core [37] yields a pronounced $A(p_{\perp})$ with an overall tilt similar to that of the experimental data and, in addition, a modulation near $p_{\perp} \approx 0.2$ au not visible in the experimental data.

To analyze the $p_{\perp}$ dependence in more detail, we now focus on the dependence of $\langle p_{\parallel} \rangle$ on $p_{\perp}$ for the two phase values $\varphi = 0$ and $\pi/2$. To this end, we plot in figure 7 $\langle p_{\parallel} \rangle$ derived from the experimental data for selected regions of the orthogonal momentum $p_{\perp}$. Each of the data points $\langle p_{\parallel} \rangle$ has been determined for slices of $p_{\perp}$ with width 0.1 au centered around the selected value of $p_{\perp}$.

We first discuss the case $\varphi = 0$ for which only one dominant wavepacket per cycle is emitted (figure 1(a)). In the absence of the Coulomb potential (e.g. in the SFCT or SFA) the part of the electron wavepacket that is created while the field increases departs directly from the parent ion on a trajectory that is commonly called a ‘direct’ one. The part of the wavepacket that is born while the field decreases is driven back to the ion by the laser electric field on a ‘recolliding’ trajectory [3]. Since Coulomb interactions are neglected in the SFCT model, both parts of the wavepacket yield $\langle p_{\parallel} \rangle = 0$ for all $p_{\perp}$. In the presence of the Coulomb interaction, the direct and recolliding portions will experience different distortion effects. While on the outgoing portion of the trajectory both parts experience a similar interaction with the Coulomb field, the recolliding portion of the wavepacket will, during its return to the parent ion, experience an additional Coulomb force. For non-zero $p_{\perp}$ it will miss the center of the binding potential and pass the ion at some distance that increases with $p_{\perp}$ [44, 45]. The additional Coulomb force is thus strongest for small $p_{\perp}$, while gating on large $p_{\perp}$ will result in reducing the influence of the Coulomb potential dominantly for the recolliding part of the emitted electron wavepackets. This trend is clearly observed for both helium and argon (figure 7(a)). The mean momentum $\langle p_{\parallel} \rangle$ gradually decreases from the large Coulomb-induced offset value at $p_{\perp} = 0$ as

New Journal of Physics 15 (2013) 043050 (http://www.njp.org/)
Figure 6. Asymmetry parameter $A(p_{\perp}, \varphi)$ characterizing the momentum spectra in the direction parallel to the laser polarization axis, $p_{\parallel}$. Solutions of the TDSE as a function of the orthogonal momentum component $p_{\perp}$ and the two-color relative phase $\varphi$ for a helium model potential [37] applying the SAE approximation (b) are compared to experimental data (c). (a) $A(p_{\perp}, \varphi)$ calculated by solving the TDSE for a short-range potential with a ground state energy of 0.5 au (see text for details) subject to a two-color pulse with half the laser peak intensity as for the helium case.

The influence of the Coulomb field on the recolliding electrons is reduced with increasing $p_{\perp}$. The SFCT (or SFA) value of $\langle p_{\parallel} \rangle = 0$ is, however, not reached, rendering, not unexpectedly, the strong-field models inapplicable for interpreting low-energy spectra. While the remaining deviation from $\langle p_{\parallel} \rangle = 0$ for large $p_{\perp}$ can be interpreted as mainly the Coulomb influence during the departure of the electrons from the parent ion, the large deviation from $\langle p_{\parallel} \rangle = 0$ for small $p_{\perp}$ can be understood by considering that an electron that is born at the peak of the laser field, without the Coulomb force, would be driven back to the ion core with zero momentum (without

New Journal of Physics 15 (2013) 043050 (http://www.njp.org/)
Figure 7. Mean value of $p_\parallel$ for helium (red) and argon (blue) as a function of $p_\perp$ for $\varphi = 0$ (a) and $\varphi = \pi/2$ (b). Open (filled) circles in (b) correspond to the high (low) momentum spectral peak observed for argon (the dip in between the two peaks is indicated by an arrow in figure 5(f)). For each data point $\langle p_\parallel \rangle$ has been calculated for slices with a width of 0.1 au centered around the respective value of $p_\perp$.

passing the origin). Thus, the driving force of such an electron due to the laser field is small and consequently the Coulomb force has a strong influence on its trajectory. The situation bears close resemblance to the case of single-cycle pulses of only one color with a carrier-envelope phase of zero. Our experimental results thus suggest that the large Coulomb-induced energy shift of photoelectrons found in simulations [24, 25] can be traced back to a strong distortion of the recolliding part of the emitted electron wavepackets.

Now we turn to the case $\varphi = \pi/2$ shown in figure 7(b), where two wavepackets per cycle are emitted (figure 1). We have discussed above that due to the influence of the Coulomb potential these two wavepackets will experience different driving forces. By gating on larger values of $p_\perp$, we dominantly reduce the effects of the Coulomb force during the recollision step and thus emphasize the relative Coulomb influence during the emission step. Gating on $p_\perp$, thus, allows one to disentangle the different Coulomb distortions to the direct and recolliding portions of the two wavepackets. For helium the mean momentum value predicted by the SFCT
model for the present laser field is 0.41. The measured value \( \langle p_\parallel \rangle \) (red squares) near \( p_\perp = 0 \) is remarkably close to this value (\( \approx 0.39 \) au). Gating on larger values of \( p_\perp \), \( \langle p_\parallel \rangle \) slightly decreases and the deviation from the SFCT prediction becomes still larger as we gradually decrease the contributions of the recolliding wavepacket portions (figure 7(b)). The measured small deviation of the spectral mean value from the SFCT value of 0.41 is thus most likely caused by stronger Coulomb distortions of the direct wavepacket portions rather than of the recolliding portions.

For argon, the experimental \( p_\parallel \) distribution features two peaks (cf figure 5(f)). Accordingly, the dependence of their positions as a function of \( p_\perp \) can be followed separately. We tentatively associate the two peaks with the direct and the recolliding wavepacket emitted during the positive and negative half-cycle, respectively (see figure 1). For the higher momentum peak, \( \langle p_\parallel \rangle \) for small \( p_\perp \) is slightly larger than the SFCT prediction. By gating on larger values of \( p_\perp \), we approach the SFCT prediction \( \approx +0.18 \), see figure 7(b). However, due to the low laser field strength used in the experiment on argon, the influence of the ionic core on the wavepacket is strong throughout its motion and a unique separation into an emission and recollision step becomes problematic. Thus, interpreting the small deviation of \( \langle p_\parallel \rangle \) at \( p_\perp = 0 \) from the SFCT as being mainly caused by the recollision step of the wavepacket, as the results seem to suggest, becomes questionable. Turning to the lower momentum peak associated with the direct emission during the positive half-cycle, we find that it remains at a value of \( \langle p_\parallel \rangle \approx -0.15 \) almost independent of \( p_\perp \), far away from the SFCT value of \( \approx +0.18 \). The insensitivity to \( p_\perp \) is expected because the wavepacket consists of only direct portions. Reducing the Coulomb influence on recolliding portions by gating on higher values of \( p_\perp \) has therefore negligible effect. The large deviation from the SFCT value demonstrates the inapplicability of SFA theories to extracting information from the direct wavepacket, especially from experiments with low intensity, where the ionic core potential plays a decisive role.

6. Wavepacket focusing by the Coulomb field

The different effect of the Coulomb field on the recolliding and direct portions of emitted wavepackets has still another consequence: figure 8(a) shows the width of the electron momentum spectrum along \( p_y \) of the helium data as a function of \( p_\parallel \). Within the adiabatic approximation to tunnel ionization, an analytic expression for the width of the electron momentum spectrum along the direction perpendicular to the polarization axis of a linearly polarized laser field due to tunnel ionization has been obtained \[41\]. Using that expression, we obtain an FWHM of 0.47 for the peak field strength of a laser pulse with \( \varphi = 0 \) used in our experiment. This value is in excellent agreement with the experimental data for \( p_\parallel > 0 \) (figure 8(a)). The experimental width for \( p_\parallel < 0 \), however, shows a pronounced deviation from the theoretical prediction (dashed line) that increases with \( |p_\parallel| \). We attribute the deviation to Coulomb focusing \[46\] of that portion of the emitted wavepacket which is driven back to the ion by the laser field and passes the Coulomb potential. During its passage it experiences a lateral force caused by the gradient of the potential that counteracts the lateral momentum spread induced during tunneling. By contrast, the portion of the wavepacket that does not pass the ion \( (p_\parallel > 0) \) is subject to only much weaker Coulomb focusing. Its momentum width is therefore close to that predicted by the tunneling theory. These findings are also in agreement with those obtained by gating on \( p_\perp \) (figure 7(a)), namely that the observed spectral asymmetry along \( p_\parallel \) is dominantly caused by the recolliding wavepacket portions. Indeed, by comparison of the measured spectrum in figure 8(b) to the SFCT spectrum shown in the same panel, it can be
Figure 8. (a) Experimentally determined width (FWHM) of the electron momentum spectrum along the perpendicular momentum coordinate, $p_y$, with $p_x$ being integrated over, for helium and $\varphi = 0$ as a function of the momentum along the laser polarization direction $p_\parallel$ (red squares). The dashed blue line shows the prediction for tunnel ionization [41]. The gray area blanks those regions where our detector has no resolution for electrons. (b) Measured ion momentum spectrum taken from figure 5(c) and mirrored about $p_\parallel = 0$ for better comparison with the electron momentum data in (a) (red line), in comparison with the SFCT spectrum (blue line).

seen that the experimentally observed spectral asymmetry in the momentum region $p_\parallel \lesssim -0.4$ overlaps with the region for which we observe the largest Coulomb focusing (compare to figure 8(a)).

7. Summary

We have investigated the influence of the Coulomb potential on the vectorial momentum distribution of electronic wavepackets ionized by strong laser pulses employing the COLTRIMS technique. Key is the use of sculpted $\omega-2\omega$ two-color pulses for which the relative phase can be tuned. This allows us to control the temporal structure of the wavepacket emission.
and propagation with sub-cycle precision. In turn, the relative importance of the laser field and Coulomb field can be controlled and modified. By comparing our measurements of the first and second moments of the momentum distributions to classical and quantum mechanical calculations with or without inclusion of the Coulomb force, we can identify the deviations from the SFCT model which is the quasi-classical limit of the SFA. We find that the direct and the recolliding parts of the wavepacket feature different levels of sensitivity to the Coulomb field. Since variants of the SFA are at the heart of our understanding of most strong-field processes as for example high-harmonic generation, e.g. [13–15], electron self-diffraction [9, 12] and electron holography [9, 47, 48], the results of our work have implications for any experiment that uses field-ionized electron wavepackets to probe the parent ion.

Acknowledgments

We acknowledge funding by the Austrian Science Fund (FWF) under grant numbers P21463-N22, SFB049-Next Lite, SFB041-VICOM and V193-N16, and by a SIRG grant from the ERC.

References

[1] Krausz F and Ivanov M 2009 Attosecond physics Rev. Mod. Phys. 81 163–234
[2] Corkum P B and Krausz F 2007 Attosecond science Nature Phys. 3 381–7
[3] Bultuška A, Ubetacker M, Goulielmakis E, Kienberger R, Yakovlev V S, Udem T, Hänisch T W and Krausz F 2003 Phase-controlled amplification of few-cycle laser pulses IEEE J. Sel. Top. Quantum Electron. 9 972–89
[4] Huang S-W et al 2011 High-energy pulse synthesis with sub-cycle waveform control for strong-field physics Nature Photon. 5 475–9
[5] Kitzler M and Lezius M 2005 Spatial control of recollision wave packets with attosecond precision Phys. Rev. Lett. 95 253001
[6] Kitzler M, Xie X, Scrinzi A and Baltuska A 2007 Optical attosecond mapping by polarization selective detection Phys. Rev. A 76 011801
[7] Kitzler M, Xie X, Roither S, Scrinzi A and Baltuska A 2008 Angular encoding in attosecond recollision New J. Phys. 10 025029
[8] Smirnova O, Mairesse Y, Patchkovskii S, Dudovich N, Villeneuve D, Corkum P and Ivanov M Yu 2004 Laser-induced interference, focusing and diffraction of rescattering molecular photoelectrons Phys. Rev. Lett. 93 223003
[9] Mairesse Y et al 2010 High harmonic spectroscopy of multichannel dynamics in strong-field ionization of diatomic molecules J. Phys. B: At. Mol. Opt. Phys. 37 L234–50
[10] Lindner F, Schätzel M, Walther H, Baltuška A, Goulielmakis E, Krausz F, Milošević D, Bauer D, Becker W and Paulus G 2005 Attosecond double-slit experiment Phys. Rev. Lett. 95 040401
[11] Yurchenko S, Patchkovskii S, Litvinyuk I, Corkum P and Yudin G 2004 Laser-induced interference, focusing and diffraction of rescattering molecular photoelectrons Phys. Rev. Lett. 93 223003
[12] Baker S, Robinson J S, Haworth C A, Teng H, Smith R A, Chirila C C, Lein M, Tisch J W G and Marangos J P 2006 Probing proton dynamics in molecules on an attosecond time scale Science 312 424–7
[13] Smirnova O, Mairesse Y, Patchkovskii S, Dudovich N, Villeneuve D, Corkum P and Ivanov M Yu 2009 High harmonic interferometry of multi-electron dynamics in molecules Nature 460 972–7
[14] Haessler S et al 2010 Attosecond imaging of molecular electronic wavepackets Nature Phys. 6 200–6
[15] Wörner H J, Bertrand J B, Kartashov D V, Corkum P B and Villeneuve D M 2010 Following a chemical reaction using high-harmonic interferometry, supplementary information Nature 466 604–7
[16] Mairesse Y et al 2010 High harmonic spectroscopy of multichannel dynamics in strong-field ionization Phys. Rev. Lett. 104 213601

New Journal of Physics 15 (2013) 043050 (http://www.njp.org/)
[18] Keldysh L V 1964 Ionization in the field of a strong electromagnetic wave Zh. Eksp. Teor. Fiz. 47 1945–57
[19] Faisal F H M 1973 Multiple absorption of laser photons by atoms J. Phys. B: At. Mol. 6 L89
[20] Reiss H 1980 Effect of an intense electromagnetic field on a weakly bound system Phys. Rev. A 22 1786–813
[21] van Linden van den Heuvel B and Muller H G 1988 Limiting cases of excess-photon ionization Proc. 4th Int. Conf. on Multiphoton Processes (JILA Boulder, CO, 13–17 July, 1987) ed S J Smith and P L Knight (Cambridge: Cambridge University Press) pp 25–34
[22] Arbó D G, Ishikawa K, Schiessl K, Persson E and Burgdörfer J 2010 Diffraction at a time grating in above-threshold ionization: the influence of the Coulomb potential Phys. Rev. A 82 043426
[23] Arbó D G, Yoshiida S, Persson E, Dimitriou K and Burgdörfer J 2006 Interference oscillations in the angular distribution of laser-ionized electrons near ionization threshold Phys. Rev. Lett. 96 143003
[24] Chelkowski S, Bandrauk A and Apolonski A 2004 Phase-dependent asymmetries in strong-field photoionization by few-cycle laser pulses Phys. Rev. A 70 013815
[25] Chelkowski S and Bandrauk A 2005 Asymmetries in strong-field photoionization by few-cycle laser pulses: kinetic-energy spectra and semiclassical explanation of the asymmetries of fast and slow electrons Phys. Rev. A 71 053815
[26] Gräfe S, Doose J and Burgdörfer J 2012 Quantum phase-space analysis of electronic rescattering dynamics in intense few-cycle laser fields J. Phys. B: At. Mol. Opt. Phys. 45 055002
[27] Smirnova O, Spanner M and Ivanov M 2008 Analytical solutions for strong field-driven atomic and molecular one- and two-electron continua and applications to strong-field problems Phys. Rev. A 77 033407
[28] Schumacher D W, Weihe F, Muller H G and Buckshaum P H 1994 Phase dependence of intense field ionization: a study using two colors Phys. Rev. Lett. 73 1344–7
[29] Muller H G, Buckshaum P H, Schumacher D W and Zavriyev A 1990 Above-threshold ionization with a two-colour laser field J. Phys. B: At. Mol. Opt. Phys. 23 2761–9
[30] Ehlotzky F 2001 Atomic phenomena in bichromatic laser fields Phys. Rep. 345 175–264
[31] Ohmura H, Nakanaga T and Tachiya M 2004 Coherent control of photofragment separation in the dissociative ionization of IBr Phys. Rev. Lett. 92 113002
[32] Ohmura H, Saito N and Tachiya M 2006 Selective ionization of oriented nonpolar molecules with asymmetric structure by phase-controlled two-color laser fields Phys. Rev. Lett. 96 173001
[33] Ray D et al 2009 Ion-energy dependence of asymmetric dissociation of D2 by a two-color laser field Phys. Rev. Lett. 103 223201
[34] De S et al 2009 Field-free orientation of CO molecules by femtosecond two-color laser fields Phys. Rev. Lett. 103 153002
[35] Xie X et al 2012 Attosecond probe of valence-electron wave packets by subcycle sculpted laser fields Phys. Rev. Lett. 108 193004
[36] Dörner R, Mergel V, Jagutzki O, Spielerberger L, Ullrich J, Moshammer R and Schmidt-Böcking H 2000 Cold target recoil ion momentum spectroscopy: a ‘momentum microscope’ to view atomic collision dynamics Phys. Rep. 330 95–192
[37] Tong X M and Lin C D 2005 Empirical formula for static field ionization rates of atoms and molecules by lasers in the barrier-suppression regime J. Phys. B: At. Mol. Opt. Phys. 38 2593–600
[38] Moshammer R et al 2000 Momentum distributions of Ne++ ions created by an intense ultrashort laser pulse Phys. Rev. Lett. 84 447–50
[39] Yudin G and Ivanov M 2001 Nonadiabatic tunnel ionization: looking inside a laser cycle Phys. Rev. A 64 013409
[40] Gopal R et al 2009 Three-dimensional momentum imaging of electron wave packet interference in few-cycle laser pulses Phys. Rev. Lett. 103 053001
[41] Delone N B and Krainov V P 1991 Energy and angular electron spectra for the tunnel ionization of atoms by strong low-frequency radiation J. Opt. Soc. Am. B 8 1207
[42] Dimitriou K, Arbó D G, Yoshiida S, Persson E and Burgdörfer J 2004 Origin of the double-peak structure in the momentum distribution of ionization of hydrogen atoms driven by strong laser fields Phys. Rev. A 70 061401

New Journal of Physics 15 (2013) 043050 (http://www.njp.org/)
[43] Lemell C, Dimitriou K, Tong X-M, Nagele S, Kartashov D, Burgdörfer J and Gräfe S 2012 Low-energy peak structure in strong-field ionization by midinfrared laser pulses: two-dimensional focusing by the atomic potential Phys. Rev. A 85 011403

[44] Dietrich P, Burnett N, Ivanov M and Corkum P 1994 High-harmonic generation and correlated two-electron multiphoton ionization with elliptically polarized light Phys. Rev. A 50 R3585–8

[45] Xie X, Scrinzi A, Wickenhauser M, Báltuška A, Barth I and Kitzler M 2008 Internal momentum state mapping using high harmonic radiation Phys. Rev. Lett. 101 033901

[46] Brabec T, Ivanov M and Corkum P 1996 Coulomb focusing in intense field atomic processes Phys. Rev. A 54 R2551–4

[47] Arbó D G, Persson E and Burgdörfer J 2006 Time double-slit interferences in strong-field tunneling ionization Phys. Rev. A 74 063407

[48] Huismans Y et al 2010 Time-resolved holography with photoelectrons Science 331 61–4