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Probing atomic and molecular targets by intense bicircular counter-rotating laser fields

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Abstract. We present an *ab initio* study of strong-field ionization of H and H_2^+ by two-colour bicircular counter-rotating 1ω - 2ω laser pulses containing 2 and 10 optical cycles at the fundamental frequency corresponding to a wavelength of 800 nm. We address effects of laser intensity, pulse duration and in the case of H_2^+ of alignment on photoelectron momentum distributions (PMDs). In the long pulse limit, as expected from a simple classical picture, the PMDs of H and of H_2^+ aligned perpendicular to the polarization plane show a three-fold symmetry imposed by the bicircular laser field. Compared with the expectation for the positions of the peaks in this three-fold structure, we observe off-set angles that depend on the photoelectron energy as well as the laser intensity. For H_2^+ , the symmetry in the PMDs is broken by the anisotropic molecular potential at molecular alignment not perpendicular to the plane of laser polarization. In the short pulse limit, the PMDs are sensitive to the carrier envelope phase of the laser field.

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1. Introduction

Strong-field ionization may take place in the tunneling regime or the multiphoton absorption regime depending on the external field strength, the binding energy of the probed electron and the wavelength of the ionizing laser as described by the Keldysh parameter [1]. In linearly polarized fields, at laser intensities around 10^{14} Wcm^{-2} and a wavelength of 800 nm, corresponding to the Ti:Sapphire laser, the dynamics is close to the tunneling regime for typical atomic and molecular targets, and the three-step

model [2, 3] has been instrumental for the development of an understanding of much of strong-field physics. In this model, after the first ionization step, the freed electron is driven by the external field in the second step and may travel directly to the detector or be driven back to the parent ion and recollide. In this case, a number of different processes may occur: The electron may scatter and gain high energy, it may excite or ionize the target or it may recombine and emit its excess energy as coherent radiation in a high-order harmonic generation (HHG) process. The electron dynamics in the laser field and the recombination process is controlled by the ellipticity and carrier envelope phase of the driving field. For example, in circularly-polarized laser fields, the laser driven electron hardly revisits its parent ion and both electron-ion rescattering and production of HHG are suppressed [4, 5]. In the case of linear polarization, where rescattering and production of HHG are substantial, both ionized and rescattered electrons move predominantly along the laser polarization axis, making it difficult to distinguish direct electrons from rescattered electrons in photoelectron momentum distributions (PMDs). It is therefore of fundamental interest to investigate the ionization dynamics for different polarizations and field configurations.

In this connection, we note the recent interest in the application of two-colour bicircular laser pulses, which can lead to production of circularly polarized HHG and attosecond pulses with circular polarization [6–9]. Such pulses have a large potential for application in, for example, investigations of circular dichroism in chiral molecules, and a number of recent studies on HHG from two-colour bicircular fields have appeared (see, e.g., references [10–15] and references therein). The focus in this paper is on strong-field ionization by bicircular two-colour counter-rotating fields. In the first combined experimental and theoretical study on argon atoms [16], the characteristic shapes of the PMDs were reported for co- and counter-rotating fields: PMDs exhibit a symmetric n -lobe structure when the combined laser fields are counter rotating, whereas a co-rotating bicircular field results in a single crescent-shaped structure in the PMDs. The number of lobes in the PMDs produced by a counter-rotating bicircular laser depends on the frequency ratio: a three-fold symmetric PMDs is obtained in the case of a 1ω - 2ω frequency combination. The significant rescattering for counter-rotating fields and its absence for co-rotating fields was also reported, as well as the decoupling of the ionization and rescattering step. Later work, also on argon [17], investigated the possibility of controlling the energy of the rescattering electron by varying the intensity ratio between the two fields. The strong-field ionization process for atoms was investigated theoretically in detail in the strong-field approximation including the rescattering term [18, 19], and more recently molecular effects were analysed within the same theoretical framework [20]. Probing of atoms and molecules by bicircular laser fields can be used to extract information about elastic scattering differential cross sections [21]. Very recently theory on strong-field photoelectron holography in atoms was reported for two-colour bicircular counter propagating fields [22]. Two-colour bicircular double ionization has also been investigated theoretically [23, 24] and experimentally, in argon atoms, control over the PMDs by the intensity ratio [25, 26] and the helicity [25]

was illustrated. Very recently experiment and S -matrix calculations addressed the role of molecular symmetry [27]. So far, the molecular case has not been investigated by TDSE calculations and it is one of the main purposes of the present paper to provide theoretical benchmark data for the simplest molecular target (the hydrogen molecular ion) for strong-field ionization in counter-rotating bicircular fields.

In this paper, we study strong-field ionization of atomic hydrogen and the molecular hydrogen ion by bicircular laser pulses. So far, strong-field studies of atoms and molecules ionized by bicircular lasers focused on PMDs obtained in the long pulse limit and explored the n -fold symmetric structure. To the best of our knowledge, no studies have been reported on atoms or molecules in bicircular laser containing few cycles, which would shed light on the effect of the carrier envelope phase. Moreover, for molecules and molecular ions, ionization by bi-circularly polarized pulses is expected to produce complicated shapes in PMDs due to the anisotropic molecular potential felt by the ionized electron [28]. Here, we study strong-field ionization of H and H_2^+ probed by bicircular laser fields with the 1ω - 2ω frequency combination by solving the three-dimensional TDSE. In particular, we focus on above-threshold ionization (ATI) spectra and PMDs and address the effects of laser intensity and pulse duration and for H_2^+ the effect of alignment. In the long pulse limit, the threefold symmetry of the ionizing field is reflected in the PMDs for H and H_2^+ aligned perpendicular to the polarization plane. We find that the PMDs are rotated by an off-set angle compared to the expectation from simple classical considerations, and we study these off-set angles as a function of photoelectron energy and laser intensity. In the molecular case, the threefold symmetry is broken when the molecular axis is not aligned perpendicular to the plane of laser polarization. In this sense, the bicircular field elucidates the symmetry of the target. In the short pulse limit, the carrier envelope phase (CEP) has substantial effect on the PMDs for both H and H_2^+ . Atomic units are used throughout unless indicated otherwise.

2. Summary of methodology and computational parameters

In this section, the methodology and the parameters used in the calculations are summarized. The method has been described in detail elsewhere [29, 30], so the discussion here will be brief. The TDSE, $i\partial_t\Psi(\vec{r}, t) = H(t)\Psi(\vec{r}, t)$, with $\Psi(\vec{r}, t)$ the wave function and $H(t)$ the time-dependent Hamiltonian is solved in the velocity gauge and dipole approximation. For the one-electron systems considered here, the Hamiltonian is given by $H(t) = -\nabla^2/2 - i\vec{A}(t) \cdot \nabla + V(\vec{r})$, where $\vec{A}(t)$ is the vector potential of the laser pulse and $V(\vec{r})$ is the atomic or molecular potential. To simplify the differential operators, it is convenient to work with the reduced wave function, $\Phi(\vec{r}, t) = r\Psi(\vec{r}, t)$. Inserting $\Psi(\vec{r}, t) = \Phi(\vec{r}, t)/r$ into the TDSE, the following equation is obtained for the reduced wave function

$$i\frac{\partial}{\partial t}\Phi(\vec{r}, t) = \left[-\frac{1}{2}\frac{\partial^2}{\partial r^2} + \frac{L^2}{2r^2} - i\vec{A}(t) \cdot \left(\nabla - \frac{\hat{\vec{r}}}{r} \right) + V(\vec{r}) \right] \Phi(\vec{r}, t), \quad (1)$$

where the first two terms on the right-hand side describe the radial and angular parts of the kinetic energy operator and the last two terms describe the interaction with the external field and atomic or molecular potential. The time-dependent reduced wave function is expanded in spherical harmonics, $Y_{lm}(\theta, \phi)$

$$\Phi(\vec{r}, t) = \sum_{l=0}^{l_{max}} \sum_{m=-l}^l f_{lm}(r, t) Y_{lm}(\theta, \phi), \quad (2)$$

which, e.g., simplifies the action of the angular momentum operator, L^2 , in equation (1). The size of the angular basis is $(l_{max} + 1)^2$, with $l_{max} = 60$ the maximum angular quantum number. We use an equidistant grid with 8192 points that extends up to 1200 au for the radial functions $f_{lm}(r, t)$. The TDSE is solved using a combined split operator Crank-Nicolson method [29, 30] with a time-step of $\delta t = 0.005$ au. The initial state is found by propagation in imaginary time. The results have been checked for convergence.

The external field of the considered counter-rotating bicircular laser pulse is defined as $\vec{F}(t) = -\partial_t \vec{A}(t)$, where $\vec{A}(t)$ is the vector potential, given by

$$\vec{A}(t) = \frac{A_0 f(t)}{\sqrt{2}} \begin{pmatrix} \cos(\omega t + \varphi) + \cos(2\omega t + \varphi) \\ \sin(\omega t + \varphi) - \sin(2\omega t + \varphi) \\ 0 \end{pmatrix}, \quad (3)$$

with A_0 the amplitude, ω the carrier angular frequency, φ the CEP, and $f(t) = \sin^2(\omega t / 2N_{cyc})$ the envelope for an N_{cyc} -cycle pulse. In the present work, we use a 1ω - 2ω frequency combination with intensity ratio of 1:1 and $\omega = 0.057$ au corresponding to 800 nm wavelength. Lissajous curves of the external fields and vector potentials for pulses containing 2 and 10 cycles are shown in figure 1. In the case $N_{cyc} = 10$ cycles, the field and the vector potential show a threefold symmetry, resulting in three ionization events or three solutions to the saddle point equation in the strong-field approximation [19, 31]. We see from the figure that a change in the CEP results in a rotation of the Lissajous curves. In the short pulse limit ($N_{cyc} = 2$ cycles), the envelope of the laser field breaks the threefold symmetry. In this case as can be seen in figure 1, changing the CEP from $-\pi/2$ to 0 rad changes the shape of the external field substantially and this will affect the resulting PMDs in a way which is not just a simple rotation of the PMD. This is contrary to circularly polarized pulses, in which case changing the CEP for spherically symmetric targets would only result in an overall rotation of the PMDs as was demonstrated in earlier studies for H and Ar atoms [32, 33].

The calculations were performed at laser peak intensity of 4.5×10^{13} Wcm $^{-2}$ with a corresponding field peak value of 0.0359 au, see figure 1(left panels). In order to investigate the effect of laser intensity on PMDs and ATI spectra, TDSE calculations were also performed for H (1s) at laser peak intensities of 2.2×10^{13} Wcm $^{-2}$ and 6.7×10^{13} Wcm $^{-2}$; all below the barrier-suppression intensity of approximately 1.4×10^{14} Wcm $^{-2}$. The calculations were performed at fixed intensity ratio of the two fields frequencies. In earlier studies [17], the shape of PMDs was found to depend on the intensity ratio of the two frequencies.

Figure 1. Lissajous curves of external fields (left panels (a), (c)) and (minus) vector potentials (right panels (b), (d)) for a 1ω - 2ω bicircular counter-rotating laser pulse containing 2 cycles (top panels (a), (b)) and 10 cycles (bottom panels (c), (d)) with a laser peak intensity $4.5 \times 10^{13} \text{ Wcm}^{-2}$, CEP values of $\varphi = -\pi/2$ (solid lines) and $\varphi = 0$ rad (dashed lines), and $\omega = 0.057$ au. The minus vector potentials in (b) and (d) give the expected final momenta from simple classical considerations (see equation (4)).

For H_2^+ , the TDSE calculations were performed at a frozen geometry with an internuclear separation of 5 au, which is larger than the equilibrium distance of 2 au. We have chosen this distance to have a clear effect of the non-spherically symmetric part of the molecular potential, while avoiding the phenomenon of enhanced ionization, which is present at larger internuclear distances due to charge resonance phenomena and coupling to the σ_u excited state [34, 35]. The ionization potential of the σ_g state at this internuclear distance is 0.729 au. The calculations were performed at laser intensities of $2.2 \times 10^{13} \text{ Wcm}^{-2}$ and $4.5 \times 10^{13} \text{ Wcm}^{-2}$ at molecular alignment angles $\beta=0$ and 90° . The alignment angle β is defined as the angle between the molecular frame and the laboratory (lab) frame z -axes, see reference [28]. In our TDSE calculations, the laser polarization is confined to the xy -plane of the lab frame (see equation (3)). At $\beta = 0^\circ$, the molecular axis is perpendicular to the plane of laser polarization. At $\beta = 90^\circ$, the molecular axis and the lab frame x -axis are aligned.

The PMDs are computed by projecting the wavepacket at the end of the laser pulse on scattering states for H [28, 36]. The scattering states were obtained by solving the radial Schrödinger equation, using the same radial grid and size of angular basis set as in the TDSE calculations. For H_2^+ , PMDs were obtained by projection on Coulomb

Figure 2. ATI (left) and PMDs (right) of H (1s) in 1ω - 2ω bicircular counter-rotating laser pulses containing 2 cycles (top) and 10 cycles (bottom panels) with a laser peak intensity $4.5 \times 10^{13} \text{ Wcm}^{-2}$, $\varphi = -\pi/2$, and $\omega = 0.057 \text{ au}$ corresponding to 800-nm wavelength. The curves in (b) and (d) show the predictions of equation (4).

scattering states in the asymptotic region [28, 36].

3. Results and discussion

This section is divided into two parts. In the first part we address effects of laser intensity and pulse duration on ATI spectra and PMDs of H. In the second part, we move on to the H_2^+ molecule, where in addition we address the effects of the nonspherical molecular potential and alignment on ATI spectra and PMDs.

3.1. H in bicircular counter-rotating laser fields: Effects of intensity and pulse duration on ATI spectra and PMDs

In figure 2, we present ATI spectra and PMDs for H, initially in the 1s state, in bicircular counter-rotating laser pulses containing 2 cycles (top panels) and 10 cycles (bottom panels), with laser peak intensities of $4.5 \times 10^{13} \text{ Wcm}^{-2}$ and a CEP of $\varphi = -\pi/2$. In the short pulse limit ($N_{cyc} = 2$), the ATI spectrum of H (1s) is characterized by a single broad peak that is rather featureless. In the long pulse limit ($N_{cyc} = 10$), the ATI spectrum contains several peaks resulting from multiphoton absorption. Increasing the number of optical cycles shifts the ATI intensity to lower photoelectron energy.

Turning to PMDs (figures 2 (b) and (d)), for short pulses ($N_{cyc} = 2$) the PMD has

Figure 3. PMDs (left column), ATI spectra (middle column) and angular distributions within each ATI peak (right column) for H (1s) at laser intensities 2.2×10^{13} (top panels), 4.5×10^{13} (middle panels), and $6.7 \times 10^{13} \text{ Wcm}^{-2}$ (bottom panels). The laser pulses contain 10 cycles. The curves in (a), (d) and (g) show the predictions of equation (4).

a general resemblance to the predicted final momentum based on the simple classical considerations shown in figure 1(b), that is

$$\vec{k} = - \int_{t_0}^{\infty} \vec{F}(t) dt = -\vec{A}(t_0), \quad (4)$$

with t_0 the time of ionization. However, a more detailed investigation of the PMD reveals the following discrepancies with equation (4): the PMD obtained from TDSE calculations is not reflection symmetric across the line $k_x = 0$ likely due to CEP effects. Moreover, the ionization peaks display an off-set (counter clockwise) angle relative to the final momentum predicted by equation (4). The off-set angle is caused by the Coulomb potential felt by the ionized electron, which changes its final momentum from the classical prediction of equation (4) [37].

For 10-cycle pulses, the electric field presented in figure 1(c) induces three main ionization events which give rise to PMDs with three-fold symmetry, in agreement with recent experiments and theory on the Ar atom [17]. The PMDs display an off-set angle compared to the final electron momentum predicted by equation (4). This off-set angle is dependent on photoelectron energy. In elliptically polarized light a dependence of the off-set angle on photoelectron energy was recently predicted for the H atom [38] and observed and modelled for the Ar atom [39]. The energy-dependent off-set angle was attributed to ionization time delays for electrons with different energies [39]. To investigate the dependence of this phenomenon on laser intensity, PMDs were computed for H at additional laser intensities of 2.2×10^{13} and $6.7 \times 10^{13} \text{ Wcm}^{-2}$, and the results

Figure 4. Intensity effect on energy-dependent off-set angle measured with respect to the expected classical peak along the k_y axis (see the 10-cycle vector potential is figure 1(d)) for H at laser intensities 2.2×10^{13} (squares), 4.5×10^{13} (circles), and $6.7 \times 10^{13} \text{ Wcm}^{-2}$ (diamonds), and H_2^+ at $\beta = 0^\circ$ alignment angle and laser intensities 2.2×10^{13} (x) and $4.5 \times 10^{13} \text{ Wcm}^{-2}$ (+).

obtained at all considered intensities are presented in figure 3. From the ATI spectra (middle column in figure 3), it is seen that the ATI peaks shift to lower energy as the laser intensity increases due to the increase in pondermotive energy. The PMDs (left column in figure 3) become more complex as the laser intensity increases, with additional progressions showing up resulting from multiphoton absorption, in addition to a low-energy structure that becomes more pronounced with increasing intensity, in particular at $6.7 \times 10^{13} \text{ Wcm}^{-2}$. The rightmost column in figure 3 includes angular distributions of photoelectrons within each ATI peak. These distributions show how the emission angle (θ_k) depends on both photoelectron energy and laser intensity, and reveal a dip in θ_k at photoelectron energy just below 0.1 au, in particular at a laser intensity $6.7 \times 10^{13} \text{ Wcm}^{-2}$.

The energy-dependent off-set angles were calculated for H from figure 3 and plotted against photoelectron energy in figure 4 at the different laser intensities. The off-set angle is defined as the difference between the expectation value of θ_k from the differential probability at a given momentum/energy value derived from the TDSE results and the expected angle of the final momentum from classical predictions based on equation (4). Since the PMDs show a threefold pattern, we select the emission peaks closest to the positive k_y axis, that is at $\theta_k \approx 90^\circ$ in figure 3 (right panels). At the higher photoelectron energies above 0.2 au, the off-set angle decreases with increasing peak intensity. This behaviour is similar to that found in argon in close-to-circularly-polarized light [40] and in naphthalene in circularly polarized light [41]. At lower photoelectron energies, however, the dependence of the off-set angle on intensity is not

Figure 5. ATI (left column) and PMDs (right column) for H_2^+ at alignment angles $\beta = 0$ (top panels) and 90° (bottom panels) and a laser intensity of $4.5 \times 10^{13} \text{ Wcm}^{-2}$. The laser pulses contain 10 cycles. The curves in (b) and (d) show the predictions of equation (4).

monotonously decreasing, a behaviour contrasting the findings for close-to-circularly-polarized and circularly polarized light [40, 41], and therefore a marker for the difference in the dynamics in these two kinds of fields.

3.2. H_2^+ in bicircular counter-rotating laser fields: Effects of intensity, pulse duration and alignment on ATI spectra and PMDs

Next, we move on to explore the PMDs originating from the σ_g ground state of aligned H_2^+ , at fixed internuclear separation of 5 au. This system provides an interesting contrast to the atomic case, due to the interplay between pulse parameters, molecular potential, alignment of the H_2^+ molecule [28, 42], and transient electron localization [43].

In figure 5, we present ATI spectra and PMDs for H_2^+ at alignment angles $\beta = 0$ and 90° . The laser pulses contain 10 cycles, and the peak laser intensity is $4.5 \times 10^{13} \text{ Wcm}^{-2}$. The energy positions of the ATI peaks in figures 5(a) and (c) depend on the alignment angle, suggesting the involvement of AC Stark shifts that depend on the alignment angle [44]. The PMD obtained at alignment angle $\beta = 0^\circ$, shown in figure 5(b), reveals a three-fold ionization pattern. These peaks reflect the three field maxima (see figure 1). The comparison between the final momenta obtained by equation (4) (full curves) and the TDSE results show that the TDSE ionization peaks have an off-set angle relative to the classical predictions. We also see that the off-set angle depends on the photoelectron energy as was the case in the H results. At $\beta = 90^\circ$, the three-fold symmetry is no

Figure 6. ATI spectra and PMDs for H_2^+ ($\beta = 0^\circ$) computed at laser intensities of $2.2 \times 10^{13} \text{ Wcm}^{-2}$ (top panels) and $4.5 \times 10^{13} \text{ Wcm}^{-2}$ (bottom panels). The curves in (b) and (d) show the predictions of equation (4). The ATI spectrum and PMDs shown in (c) and (d) are the same as those in figure 5(a) and (b).

longer imprinted in the PMD: the ionization probability is substantially suppressed at $\theta_k \approx -45^\circ$. The symmetry in the polarization plane is lost due to the anisotropic molecular potential and possible coupling to excited states [42].

To address the effect of laser intensity on the energy-dependent off-set angle, PMDs were computed for H_2^+ at a lower laser intensity of $2.2 \times 10^{13} \text{ Wcm}^{-2}$. The calculations were performed at alignment angle $\beta = 0^\circ$ in order to eliminate any effects caused by the anisotropic molecular potential and coupling to excited states. The resulting ATI spectra and PMDs are presented in figure 6. The ATI spectrum includes a new feature, namely a low-energy shoulder on the ATI peaks at energy around 0.05 au. This shoulder can also be seen in the ATI rings in the PMD of figure 6(b), but the exact origin of this feature is presently not identified. Regarding the PMDs, three-fold symmetries are observed at both intensities and energy and intensity dependent off-set angles are observed. To see clearly the dependence on energy and intensity, the off-set angles are plotted for H_2^+ in figure 4 at laser intensities $2.2 \times 10^{13} \text{ Wcm}^{-2}$ and $4.5 \times 10^{13} \text{ Wcm}^{-2}$. The off-set angle decreases with increasing laser intensity, as is the case for close-to-circularly [40] and circularly [41] polarized light. Compared to the H results of figure 4, the H_2^+ results show monotonic behaviour, which highlights the sensitivity of the off-set angle on the atomic or molecular potential.

To address CEP effects, TDSE calculations were carried out for H_2^+ at alignment angles $\beta = 0$ and 90° and at CEP values of $-\pi/2$ and 0 rad. The laser pulses contain

Figure 7. (a,d) ATI spectra and (b,c,e,f) PMDs for H_2^+ in 1ω - 2ω bicircular counter-rotating laser fields at (b) $\beta = 0^\circ$ and CEP $\varphi = -\pi/2$ (solid line in (a)), (c) $\beta = 90^\circ$ and CEP $\varphi = 0$ (dotted line in (a)), (e) $\beta = 90^\circ$ and CEP $\varphi = -\pi/2$ (dashed line in (d)), and (f) $\beta = 90^\circ$ and CEP $\varphi = 0$ (dashed line in (d)). The laser pulses contain 2 cycles, and the peak intensity is $4.5 \times 10^{13} \text{ Wcm}^{-2}$. The curves in (b), (c), (e) and (f) show the predictions of equation (4).

2 cycles and have a peak intensity of $4.5 \times 10^{13} \text{ Wcm}^{-2}$. The resulting ATI spectra and PMDs are shown in figure 7. The ATI spectra in figure 7(a) and (d) reveal larger ionization yield in the in-polarization-plane geometry ($\beta = 90^\circ$), compared to the out-of-polarization-plane geometry ($\beta = 0^\circ$). The CEP has more effect on ATI spectra at the ionization peak at $\beta = 0^\circ$ compared to $\beta = 90^\circ$. Turning to PMDs, the CEP effect is clearly reflected in the PMDs. At $\beta = 0^\circ$, the molecular potential is rotationally symmetric in the plane of laser polarization, making the interpretation of CEP effect more straightforward. In this case, changing the CEP from $-\pi/2$ to 0 rad changes the shape of the external field (see figure 1), thereby creating a substantially different PMDs as can be seen in figure 7(b) and (c). At CEP= $-\pi/2$, the PMD in figure 7(b) contains 2 ionization peaks at emission angles $\theta_k \approx -50^\circ$ and -180° at corresponding photoelectron energies of $\approx 0.2 \text{ au}$ and 0.1 au , respectively. At CEP=0 rad, the PMD in figure 7(c) contains only a single ionization peak at $\theta_k \approx -30^\circ$ and corresponding photoelectron energy slightly above 0.1 au . These results make an interesting contrast to PMDs of H_2^+ in circularly polarized laser, in which case changing the CEP from $-\pi/2$ to 0 rad would result in an overall rotation of the PMD [32, 33, 42]. At $\beta = 90^\circ$, the molecular potential of H_2^+ is strongly anisotropic in the plane of the laser polarization, with substantial impact on the resulting PMDs: namely that the emission peaks are significantly narrower and the emission angle are different. At CEP = $-\pi/2$, the distribution at $\beta = 90^\circ$ has two ionization peaks at $\theta_k \approx -170^\circ$ and -130° (figure 7(e)). At this alignment angle, changing the CEP from $-\pi/2$ to 0 rad results in suppressing the ionization peak at -130° and the main ionization peak is found at emission angle

$\theta_k \approx 180^\circ$ (figure 7(f)). This is in contrast to a final emission angle of 0° from classical predictions based on equation (4).

4. Conclusions

In this study of strong-field ionization of atomic and molecular targets by two-colour bicircular counter-rotating laser pulses, we focused attention on ATI spectra and PMDs. The three-dimensional TDSE was solved and ATI spectra and PMDs were obtained by projecting the wavepacket on scattering states, or Coulomb waves in the asymptotic region.

For H and for H_2^+ aligned perpendicular to the polarization plane probed by 10-cycle pulses, the PMDs revealed a three-fold symmetry in the PMDs. Compared with a simple classical estimate, the PMD is rotated by an off-set angle that depends on photoelectron energy and laser intensity. The distinct peaks in the three-peaked PMDs obtained by two-colour bicircular counter-rotating field in the many-cycle limit ($N_{cyc} \gtrsim 10$) and their shifting with photoelectron energy and intensity seems a suitable observable for addressing questions of energy-dependent continuum wave phase shifts and time-delays in strong-field ionization [39]. The symmetry in the PMDs is broken as the short pulse limit is reached due to CEP effects. For H_2^+ , the threefold symmetry of PMDs depends on molecular alignment due to the anisotropic molecular potential: the three-fold symmetric PMD is obtained when the molecular axis is aligned perpendicular to the plane of laser polarization, because of rotation symmetry of molecular potential in that plane. The symmetry in the PMD is broken when the molecule is aligned parallel to the plane of laser polarization. In this sense the presence or absence of symmetry patterns in the PMDs following strong-field ionization by two-colour bicircular counter-rotating fields is a direct marker of the presence or absence of rotational symmetry in the potential felt by the active electron.

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References

- [1] Keldysh L V 1965 Ionization in the field of a strong electromagnetic wave *Sov. Phys. JETP* **20** 1307–1314.
- [2] Krause J L, Schafer K J, and Kulander K C 1992 High-Order Harmonic Generation from Atoms and Ions in the High Intensity Regime *Phys. Rev. Lett.* **68** 3535.
- [3] Corkum P B 1993 Plasma Perspective on Strong Field Multiphoton Ionization *Phys. Rev. Lett.* **71** 1994.
- [4] Hickstein D D *et al* 2012 Direct Visualization of Laser-Driven Electron Multiple Scattering and Tunneling Distance in Strong-Field Ionization *Phys. Rev. Lett.* **109** 073004.

- [5] Abu-samha M and Madsen L B 2011 Interrogation of orbital structure by elliptically polarized intense femtosecond laser pulses *Phys. Rev. A* **84** 023411.
- [6] Fleischer A, Kfir O, Diskin T, Sidorenko P and Cohen O 2014 Spin angular momentum and tunable polarization in high-harmonic generation. *Nat. Photon.* **8** 543–549.
- [7] Kfir O, Grychtol P, Turgut E, Knut R, Zusin D, Popmintchev D, Popmintchev T, Nembach H, Shaw J M, Fleischer A, Kapteyn H, Murnane M, and Cohen O 2015 Generation of bright phase-matched circularly-polarized extreme ultraviolet high harmonics *Nat. Photon.* **9** 99.
- [8] Kfir O, Grychtol P, Turgut E, Knut R, Zusin D, Fleischer A, Bordo E, Fan T, Popmintchev D, Popmintchev T, Kapteyn H, Murnane M, and Cohen O 2016 Helicity-selective phase-matching and quasi-phase matching of circularly polarized high-order harmonics: towards chiral attosecond pulses *J. Phys. B: At. Mol. Opt. Phys.* **49** 123501.
- [9] Hickstein D D, Dollar F J, Grychtol P, Ellis J L, Knut R, Hernández-García C, Zusin D, Gentry G, Shaw J M, Fan T, Dorney K M, Becker A, Jaroń-Becker A, Kapteyn H C, Murnane M M, and Durfee C G 2015 Non-collinear generation of angularly isolated circularly polarized high harmonics *Nat. Photon.* **9** 743.
- [10] Odžak S, Hasović E, and Milošević D B 2016 High-order harmonic generation in polyatomic molecules induced by a bicircular laser field. *Phys. Rev. A* **94** 033419.
- [11] Baykusheva D, Brennecke S, Lein M, and Wörner H J 2017 Signatures of electronic structure in bicircular high-harmonic spectroscopy. *Phys. Rev. Lett.* **119** 203201.
- [12] Reich D M and Madsen L B 2016 Rotating-frame perspective on high-order-harmonic generation of circularly polarized light. *Phys. Rev. A* **93** 043411.
- [13] Reich D M and Madsen L B 2016 Illuminating molecular symmetries with bicircular high-order-harmonic generation. *Phys. Rev. Lett.* **117** 133902.
- [14] Kevin M. Dorney K M *et al.* 2017 Helicity-selective enhancement and polarization control of attosecond high harmonic waveforms driven by bichromatic circularly polarized laser fields. *Phys. Rev. Lett.* **119** 063201.
- [15] Bandrauk A D and Yuan K J 2018 Molecular electron recollision dynamics in intense circularly polarized laser pulses. *J. Phys. B: At. Mol. Opt. Phys.* **51** 074001
- [16] Mancuso C A *et al.* 2015 Strong-field ionization with two-color circularly polarized laser fields. *Phys. Rev. A* **91** 031402.
- [17] Mancuso C A *et al.* 2016 Controlling electron-ion rescattering in two-color circularly polarized femtosecond laser fields. *Phys. Rev. A* **93** 053406.
- [18] Hasović E and Becker W and Milošević D B 2016 Electron rescattering in a bicircular laser field. *Opt. Express* **24** 6413–6424
- [19] Milošević D B and Becker W 2016 Improved strong-field approximation and quantum-orbit theory: Application to ionization by a bicircular laser field. *Phys. Rev. A* **93** 063418.
- [20] Busuladžić M, Gazibegović-Busuladžić A, and Milošević D B 2017 Strong-field ionization of homonuclear diatomic molecules by a bicircular laser field: Rotational and reflection symmetries. *Phys. Rev. A* **95** 033411.
- [21] Hoang V H, Le V H, Lin C D, and Le A-T 2017 Retrieval of target structure information from laser-induced photoelectrons by few-cycle bicircular laser fields. *Phys. Rev. A* **95** 031402.
- [22] Li M, Jiang W-C, Xie H, Luo S, Zhou Y and Lu P 2018 Strong-field photoelectron holography of atoms by bicircular two-color laser pulses *Phys. Rev. A* **97** 023415.
- [23] Chaloupka J L and Hickstein D D 2016 Dynamics of Strong-Field Double Ionization in Two-Color Counterrotating Fields. *Phys. Rev. Lett.* **116** 143005
- [24] Xu T T, Zhu Q Y, Chen J H, Ben S, Zhang J, and Liu X S 2018 Multiple recollisions in nonsequential double ionization by counter-rotating two-color circularly polarized laser fields. *Opt. Express* **26** 1645–1654.
- [25] Mancuso C A *et al.* 2016 Controlling nonsequential double ionization in two-color circularly polarized femtosecond laser fields. *Phys. Rev. Lett.* **117** 133201.
- [26] Eckart S *et al.* 2016 Nonsequential double ionization by counterrotating circularly polarized two-

- color laser fields. *Phys. Rev. Lett.* **117** 133202.
- [27] Lin K *et al.* 2017 Comparison Study of Strong-Field Ionization of Molecules and Atoms by Bicircular Two-Color Femtosecond Laser Pulses. *Phys. Rev. Lett.* **119** 203202.
- [28] Abu-samha M and Madsen L B 2016 Alignment dependence of photoelectron momentum distributions of atomic and molecular targets probed by few-cycle circularly polarized laser pulses. *Phys. Rev. A* **94** 023414.
- [29] Kjeldsen T K, Nikolopoulos L A A, and Madsen L B 2007 Solving the m-mixing problem for the three-dimensional time-dependent Schrödinger equation by rotations: Application to strong-field ionization of H_2^+ . *Phys. Rev. A* **75** 063427.
- [30] Kjeldsen T K 2007 Wave packet dynamics studied by ab initio methods: Applications to strong-field ionization of atoms and molecules PhD thesis, Aarhus University, Denmark
- [31] Hasović E, Kramo A, and Milošević D B 2008 Energy- and angle-resolved photoelectron spectra of above-threshold ionization and detachment. *Eur. Phys. J. Special Topics* **160** 205–216.
- [32] Martiny C P J, Abu-samha M and Madsen L B 2010 Ionization of oriented targets by intense circularly polarized laser pulses: Imprints of orbital angular nodes in the two-dimensional momentum distribution *Phys. Rev. A* **81** 063418.
- [33] Martiny C P J and Madsen L B 2006 Symmetry of Carrier-Envelope Phase Difference Effects in Strong-Field, Few-Cycle Ionization of Atoms and Molecules *Phys. Rev. Lett.* **97** 093001.
- [34] Zuo Y and Bandrauk A D 1995 Charge-resonance- enhanced ionization of diatomic molecular ions by intense lasers. *Phys. Rev. A* **52** R2511.
- [35] Chu X and Chu S-I 2000 Complex-scaling generalized pseudospectral method for quasienergy resonance states in two-center systems: Application to the Floquet study of charge resonance enhanced multiphoton ionization of molecular ions in intense low-frequency laser fields *Phys Rev A* **63** 013414.
- [36] Madsen L B, Nikolopoulos L A A, Kjeldsen T K, and Fernandez J 2007 Extracting continuum information from $\psi(t)$ in time-dependent wave-packet calculations. *Phys. Rev. A* **76** 063407.
- [37] Martiny C P J, Abu-samha M and Madsen L B 2009 Counterintuitive angular shifts in the photoelectron momentum distribution for atoms in strong few-cycle circularly polarized laser pulses. *J. Phys. B: At. Mol. Opt. Phys.* **42** 161001.
- [38] Murakami M and Chu Shih-I 2016 Photoelectron momentum distributions of the hydrogen atom driven by multicycle elliptically polarized laser pulses. *Phys. Rev. A* **93** 023425
- [39] Xie H, Li M, Luo S, Li Y, Zhou Y, Cao W, and Lu P 2017 Energy-dependent angular shifts in the photoelectron momentum distribution for atoms in elliptically polarized laser pulses. *Phys. Rev. A* **96** 063421.
- [40] Pfeiffer A N, Cirelli C, Smolarski M, Dimitrovski D, Abu-samha M, Madsen L B, and Keller U 2012 Attoclock reveals natural coordinates of the laser-induced tunnelling current flow in atoms *Nature Physics* **8** 76–80.
- [41] Maurer J, Dimitrovski D, Christensen L, Madsen L B, and Stapelfeldt H 2012 Molecular-Frame 3D Photoelectron Momentum Distributions by Tomographic Reconstruction *Phys. Rev. Lett.* **109** 123001.
- [42] Abu-samha M and Madsen L B 2017 Interplay between pulse parameters and alignment of molecular hydrogen. *J Phys.: Conf. Series* **869** 012009.
- [43] Miller M R, Jaron-Becker A and Becker A 2017 Signatures of transient electron localisation in high-order harmonic generation. *Mol. Phys.* **115** 1758–1767.
- [44] Chu Xi 2008 Molecular-orientation-dependent ac stark effect and its impact on multiphoton processes. *Phys. Rev. A* **78** 043408.