Nonsequential double ionization with polarization-gated pulses

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Abstract

We investigate laser-induced nonsequential double ionization by a polarization-gated laser pulse, constructed by employing two counter-rotating circularly polarized few-cycle pulses with a time delay \(T_d\). We address the problem within a classical framework and mimic the behaviour of the quantum-mechanical electronic wave packet by means of an ensemble of classical electron trajectories. These trajectories are initially weighted with the quasi-static tunnelling rate and with suitably chosen distributions for the momentum components parallel and perpendicular to the laser-field polarization in the temporal region for which it is nearly linearly polarized. We show that, if the time delay \(T_d\) is of the order of the pulse length, the electron-momentum distributions, as functions of the parallel momentum components, are highly asymmetric and dependent on the carrier-envelope (CE) phase. As this delay is decreased, this asymmetry gradually vanishes. We explain this behaviour in terms of the available phase space, the quasi-static tunnelling rate and the recollision rate for the first electron for different sets of trajectories. Our results show that the polarization-gating technique may provide an efficient way to study the NSDI dynamics in the single-cycle limit without employing few-cycle pulses.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Polarization gating, since proposed in the mid-1990s [1], has become a powerful tool for attosecond metrology and dynamic imaging of matter [2], mainly due to its simplicity and reliability. The key idea behind it is to combine laser fields of different polarizations and sometimes frequencies in such a way as to obtain controllable, near-linearly polarized, extremely short driving laser pulses. An important application of this technique is, for instance, the generation of isolated attosecond pulses [3–5], some of them as short as 130 attoseconds [6]. The latter pulses have been accomplished by employing, as a driving field, a left circularly polarized and a time-delayed right circularly polarized sub 10 fs laser pulse. The resulting driving pulse changes from circularly polarized to almost linearly polarized and then back to circularly polarized within the pulse envelope.

The importance of these drastic ellipticity changes is a consequence of the physical mechanism behind high-order harmonic generation (HHG). This phenomenon, which is employed in the generation of attosecond pulses, is the result of a three-step physical process [7]. Thereby, the valence electron of an atom, subjected to an intense laser field, is released in the continuum by tunnel ionization. This electron is then accelerated by the field and, depending on its energy, ionization time and on the laser-field polarization, it may be driven back towards its parent ion and recombine at a later instant. Upon recombination, the energy acquired by the electron from the field is released as a high-frequency photon. Gating is important, as it may be employed to restrict the electron ionization and recombination times to very narrow intervals. If the field is circularly polarized, the returning electron will miss the atomic core, whereas if the field is linearly polarized, the electron will interact with it.

Laser-induced nonsequential double ionization (NSDI) is caused by a similar mechanism. The main difference lies in the fact that the returning electron, instead of recombining with a bound state of its parent ion, will rescatter inelastically
with it, giving part of its kinetic energy to release a second electron. This nonsequential physical mechanism has been revealed by very peculiar features in the electron-momentum distributions, namely symmetric peaks at the nonvanishing momenta \( p_{1i} = p_{2i} = \pm 2\sqrt{U_p} \), where \( U_p \) denotes the ponderomotive energy and \( p_{ni}(n = 1, 2) \) denotes the electron-momentum components parallel to the laser-field polarization [8]. Therefore, we expect that NSDI may also be strongly influenced by the time dependence of the external laser field, for example, by the dramatic change of its amplitude and polarization. This is supported by our previous work on NSDI with few-cycle, linearly polarized driving pulses [9–11]. Therein, we have shown, both theoretically [10, 11] and experimentally [9], that the NSDI dynamics are strongly dependent on the carrier-envelope (CE) phase. Indeed, we observed strongly asymmetric distributions, which would shift from the positive to the negative region, or vice versa, of the plane \( (p_{1i}, p_{2i}) \) spanned by the electron-momentum components \( p_{ni}(n = 1, 2) \) parallel to the laser-field polarization, as the CE phase was changed. Furthermore, it has been shown, for NSDI with a single elliptically polarized driving field, that the electron-momentum distributions are highly dependent on the driving-field polarization [12].

Hence, one can raise the question of whether polarization gating may be used to control the NSDI dynamics. This issue is of interest for two main reasons. First, since one may build an extremely short, almost linearly polarized driving field, it may be possible to steer the electron motion with a much higher precision than with, for instance, linearly polarized few-cycle pulses. Second, sometimes the changes observed in the NSDI electron-momentum distributions, with regard to the driving-field shape, are far more dramatic than those in higher-order harmonic or above-threshold ionization spectra. This is due to the fact that different sets of electron orbits may be mapped into different momentum regions. Apart from that, if the second electron is dislodged by electron-impact ionization, there will be a minimum and a maximum parallel momentum for which this process is classically allowed. Depending on the field parameters, we can employ this particular feature to make whole momentum regions appear or collapse. The above-stated effects have been observed for NSDI with linearly polarized few-cycle pulses and led to dramatic changes in the electron-momentum distributions [9–11].

In this work, we investigate differential electron-momentum distributions in NSDI with polarization gating. For this purpose, we extend the classical model employed in our previous publications [10, 11] to the elliptical-polarization case. In this model, the quantum-mechanical transition probability corresponding to the scenario in which the second electron is released by electron-impact ionization, within the strong-field approximation, is mimicked by a classical ensemble of electrons released in the continuum with a quasi-static tunnelling rate [10, 11, 13]. The main difference is that, for elliptically polarized fields, one must take into account that the lateral residual laser electric field influences the electron orbits in the continuum and also at the instant of ionization. For the external driving field, we consider the same pulse configuration as in [5], i.e., two counter-rotating, time-delayed circularly polarized few-cycle pulses. We investigate the influence of both the CE phase and the delay between the two pulses on the electron-momentum distributions. In a more general context, it is worth mentioning that classical models have proven to be very powerful in the context of nonsequential double [14] or multiple [15] ionization.

This paper is organized as follows. In section 2, we provide a brief discussion of our model, placing a particular emphasis on how it differs from its counterpart for linearly polarized fields. Subsequently (section 3), we present the differential electron-momentum distributions and analyse their main features in terms of electron trajectories. Finally, our conclusions and a summary of this work are given in section 4.

2. Model

We consider an electron ensemble subject to a pulse with a time-dependent ellipticity, which is generated by the superposition of a left-circularly polarized pulse and right-circularly polarized pulse. The two pulses are taken to be identical except for their polarization, and there is a time delay \( T_d \) between them. Below, we provide more details about the pulse shape, and our classical ensemble model, which is employed to imitate the behaviour of a quantum-mechanical wave packet. We use atomic units throughout.

2.1. Polarization-gated driving field

Explicitly, the electric fields \( \vec{E}_l(t) \) and \( \vec{E}_r(t) \) of the left- and right-circularly polarized pulses read

\[
\vec{E}_l(t) = E_0 e^{-2\ln(2)(t-T_d/2)/\tau_p} \left[ \hat{\chi} \cos(\omega(t - T_d/2) + \phi) + \hat{\gamma} \sin(\omega(t - T_d/2) + \phi) \right]
\]

and

\[
\vec{E}_r(t) = E_0 e^{-2\ln(2)(t+T_d/2)/\tau_p} \left[ \hat{\chi} \cos(\omega(t + T_d/2) + \phi) - \hat{\gamma} \sin(\omega(t + T_d/2) + \phi) \right],
\]

respectively. In the above-stated equations, \( E_0 \) is the peak-field amplitude, \( \omega \) is the carrier frequency, \( \tau_p \) is the pulse duration, \( T_d \) is the time delay between the two circularly polarized pulses and \( \phi \) is the CE phase. The unit vectors in the \( x \)- and \( y \)-directions are denoted by \( \hat{\chi} \) and \( \hat{\gamma} \).

The electric field components of the combined laser pulse in the \( x \)- and \( y \)-directions are given by

\[
\vec{E}_x(t) = E_0 e^{-2\ln(2)(t-T_d/2)/\tau_p} \left[ \cos(\omega(t - T_d/2) + \phi) \right] + E_0 e^{-2\ln(2)(t+T_d/2)/\tau_p} \left[ \cos(\omega(t + T_d/2) + \phi) \right]
\]

and

\[
\vec{E}_y(t) = E_0 e^{-2\ln(2)(t-T_d/2)/\tau_p} \left[ \sin(\omega(t - T_d/2) + \phi) \right] - E_0 e^{-2\ln(2)(t+T_d/2)/\tau_p} \left[ \sin(\omega(t + T_d/2) + \phi) \right],
\]

respectively. The time-dependent ellipticity of this pulse is

\[
\xi(t) = \left[ 1 - \exp \left[ -4 \ln(2) T_d/\tau_p^2 \right] \right] / \left[ 1 + \exp \left[ -4 \ln(2) T_d/\tau_p^2 \right] \right].
\]

In the vicinity of \( t = 0 \), \( \xi(t) \) increases from 0 to 0.2 and is approximately linear. Outside this interval, this
approximation does not hold. The temporal region for which the field polarization is approximately linear is known as the ‘polarization gate’ [1].

In [5], the polarization gate for the specific laser-field configuration discussed above has been estimated to be around 0.3τp. Therein, it has also been shown that it is inversely proportional to the time delay \( T_d \) between the two pulses. Hence, to reduce the polarization gate, we can either use shorter circularly polarized pulses or increase \( T_d \). From the experimental perspective, either of them can be controlled at will. In fact, the length of the laser pulse will be conditioned by the state-of-the-art ultrafast laser technique [16]. However, the delay time between the two circularly polarized laser pulses cannot be too long. If we use a too long delay between the two circularly polarized laser pulses to obtain a sufficiently intense combined pulse, it may be necessary to strengthen our laser pulses so much that the ground-state sample may be depleted and result in a poor signal-to-noise ratio.

2.2. Electron-momentum distributions

We will now discuss how we mimic the quantum-mechanical electron-momentum distributions in a classical framework. Similar models have been employed in [10, 11, 13] for linearly polarized fields. We consider a set of classical trajectories, starting at different tunnelling times \( t_0 \) throughout the pulse. We limit such times to the time interval for which the field is almost linearly polarized. This is justified, as, only in this time range, a significant contribution to NSDI will occur. Furthermore, in the specific model discussed in this paper, we make three main assumptions. First, each trajectory is weighted with the tunnelling probability per unit time given by the well-known quasi-static formula [17]

\[
W_{t}(t_0) \sim \frac{1}{|E_{sum}(t_0)|} \exp \left[ -\frac{2(2|E_{IP1}|)^{3/2}}{3|E_{sum}(t_0)|} \right],
\]

where \( E_{IP1} \) is the first ionization threshold of the atom in question and \( E_{sum}(t_0) = E_x(t_0) + E_y(t_0) \) is the electric field of the combined, polarization-gated laser pulse. Second, to simulate the initial wave-packet spreading, an initial lateral velocity \( v_l \) is further introduced. Each trajectory is then weighted with the tunnelling probability times the quantum-mechanical transverse velocity distribution weight [18], \( W_l \), which can be calculated by

\[
W_l(v_l) = \frac{1}{\pi(\delta v_l)^2} \exp \left[ -\left( \frac{v_l}{\delta v_l} \right)^2 \right],
\]

where the lateral velocity width is given by

\[
\delta v_l = \frac{1}{E_{sum}/\sqrt{2E_{IP1}}}^{1/2}.
\]

The range for the transverse velocity distribution chosen here is \( 2\delta v_l \). Finally, a quantum-mechanical wave packet also spreads in time so that the contributions of the longer orbits are weakened. We incorporate this final ingredient in a similar way as in [12] by introducing an extra weight \((t_1 - t_0)^{-3}\) to each electron trajectory. Thereby, \( t_1 \) denotes the time the electron returned to the core. Therefore, the overall weight of each trajectory in the ensemble reads

\[
W_k(t_0, v_l) = W_l(v_l) \times W_l(v_l) \times (t - t_0)^{-3}.
\]

After tunnelling, the equations of motion for the electron in both directions are

\[
\dot{x} = \vec{E}_x(t),
\]

\[
\dot{y} = \vec{E}_y(t).
\]

In order to obtain the electron orbits, equations (10) and (11) are integrated, and the Coulomb potential is ignored. This is a reasonable assumption for a strong driving field. Electrons are assumed to be ‘born’ at time \( t_0 \) at the origin \( x = 0, y = 0 \) with an initial lateral velocity. Only those electrons coming back to the core with energy larger than \( E_{IP2} \), the second ionization threshold of the atom, will contribute to the NSDI yield. This corresponds to the physical situation in which the first electron dislodges the second by electron-impact ionization. After a trajectory is launched, by checking the position of the electron, we determine whether it has returned to the core, and, if so, its return time. The electron velocities in both directions, \( \dot{x} \) and \( \dot{y} \), are then evaluated. The energy of the electron, upon return, is

\[
E_{ret} = \frac{1}{2}(x^2 + y^2).
\]

If the condition \( E_{ret} > E_{IP2} \) is satisfied [13], the first electron gives part of its kinetic energy \( E_{ret} \) upon return to the second electron, so that it is able to overcome the second ionization threshold \( E_{IP2} \). The electron pair then obeys

\[
\frac{1}{2} \sum_{j=1}^{2} (p_{jx} + A_x(t_1))^2 = E_{ret} - E_{IP2} - \frac{1}{2} \sum_{j=1}^{2} p_{jz}^2 - \frac{1}{2} \sum_{j=1}^{2} (p_{jy} + A_y(t_1))^2.
\]

In equation (13), \( A_x(t) \) and \( A_y(t) \) denote the vector potential components

\[
A_x(t) = - \int E_x(t) \, dt
\]

and

\[
A_y(t) = - \int E_y(t) \, dt
\]

of the laser pulse in the \( x- \) and \( y- \) directions, and \( p_{jx}, p_{jy}, \) and \( p_{jz} \) are the final momentum components recorded by the detector in the \( x-, y-, \) and \( z- \) directions after tunnelling.

One should note that the energy-conservation condition (equation (13)) gives the equation of a hypersphere in the six-dimensional space spanned by the momentum components of the two electrons. This hypersphere exhibits the radius \([2(E_{ret}(t_1) - E_{IP2})]^{1/2}\) and is centred at \((p_{1x}, p_{1y}, p_{1z}, p_{2x}, p_{2y}, p_{2z}) = (A_x(t_1), -A_x(t_1), 0, -A_y(t_1), A_y(t_1), 0)\). Therefore, the larger the electron kinetic energy, the larger the region for which electron-impact ionization is classically allowed. Furthermore, the above-stated equation shows that the distributions should be centred at nonvanishing electron momenta. Specifically for monochromatic, linearly polarized fields, the vector potential upon return can be approximated by \(2\sqrt{U_p}\). This corresponds to the situation in which the electron leaves at a field maximum and returns at a crossing. Since this is the most probable momentum for
the electron upon return, we expect the distributions to be centred at this quantity. We have observed however, for the parameters employed in this work, that this estimate roughly holds.

The electron-momentum distributions then read

\[
R \sim \iint \delta(d_0 \sin^2 W_\perp(t_0, \tau)) |V_{p[n,k]}|^2 \times \delta(E_{\text{rel}}(t_1) - E_{IP2} - \frac{2}{2} \frac{p_{jx} + A_x(t_1)}{2} - \frac{2}{2} \frac{p_{jy} + A_y(t_1)}{2}) \text{.} \tag{16}
\]

The argument of the \(\delta\)-function gives the energy-conservation restriction. Since the situation addressed in this paper occurs only in the \(xy\)-plane, the electron motion in the \(z\)-direction is ignored. The form factor \(V_{p[n,k]}\) depends on the final momenta \(p_n(n = 1, 2)\) and on the intermediate momentum \(k\) in the above distributions. This term is a signature of the interaction \(V_{12}\) by which the second electron is dislodged. For instance, for a Coulomb-type interaction \(V_{12} = 1/|r_1 - r_2|\) the form factor is given by

\[
V_{p[n,k]} \sim \frac{1}{|p_1 - k|^2} \left( \frac{1}{|p_1 + p_2 - k + A(t_1)|^2 + 2E_{IP2}^2} \right) \text{.} \tag{17}
\]

This form factor favours unequal momenta. If, on the other hand, we assume that the second electron is released by a contact-type interaction \(V_{12} = \delta(r_1 - r_2)\delta(r_2)\) placed at the position of the ion, \(V_{p[n,k]} \sim \text{const.}\) and the momentum distributions are mainly determined by the momentum-space integration (for details see [13]). Therefore, such distributions are isotropic in momentum space, have the radius of the above-mentioned hypersphere and are centred at the most probable momentum upon return. This guarantees that we investigate only the effects of the gating on the NSDI momentum distributions, and thus make the polarization-gating effects clearer. It should be noted that, though the electron-momentum distribution is to some extent altered by adopting the Coulomb-type interaction, the effects related to gating discussed in this paper are not influenced, as demonstrated later. In the following, we will employ mostly the contact-type interaction, unless explicitly stated.

### 3. Results

In this section, we display the NSDI electron-momentum distributions computed with equation (16) and the polarization-gated pulse described in section 2.1. This pulse has been chosen as the superposition of two time-delayed, circularly polarized pulses of wavelength \(\lambda = 800\text{ nm}\). The full width at half maximum (FWHM) of the circularly polarized light is chosen as four cycles, i.e., \(\tau \sim 10\text{ fs}\). The distributions were calculated for neon. For this specific species, the dominant physical mechanism is electron-impact ionization. For other species, such as argon, the excitation of the parent ion by the returning electron, with subsequent double ionization, plays an important role [8, 19].

Such distributions are depicted in figure 1, as functions of the electron-momentum components parallel to the laser-field polarization at the very centre of the gate. For the pulse shape considered in this paper, this corresponds to the \(x\)-direction. Unless otherwise stated (see, e.g., figures 5 and 6), we will denote such momentum components as \(p_{n||}(n = 1, 2)\). One should note, however, that in the calculations, the electron is propagating in both \(x\)- and \(y\)-directions.

If the delay phase between both pulses is equal to the pulse length, i.e. \(\omega T_d = 8\pi\), the distributions are, in general, highly asymmetric and concentrated either in the positive or in the negative parallel momentum regions. For the specific parameters in this work, as the CE phase \(\phi\) is changed from \(\phi = 0.5\tau\) to \(\phi = 1.2\tau\), the distributions shift from the third to the first quadrant of the plane \((p_{1||}, p_{2||})\) spanned by the
parallel-momentum components. This behaviour is shown in the far left panels of figure 1, i.e. in figures 1(a), (e), (i) and (m), and resembles to a great extent what happens for linearly polarized few-cycle pulses.

In this latter case, we have shown that the asymmetry and the shifts in the distributions were due to the changes in the dominant set of trajectories along which the first electron would return. This set of trajectories strongly depends on the CE phase, so that the asymmetry can be used to determine this parameter [10, 11]. The behaviour of the electron-momentum distributions in the present case suggests a similar physical interpretation. From the positions of the maxima, at approximately \(2\sqrt{U_p}, 2\sqrt{U_p}\) and \((-2\sqrt{U_p}, -2\sqrt{U_p})\), we verify that the estimates for the peaks of the distributions, for monochromatic, linearly polarized fields, roughly hold in this case.

As the delay phase is reduced to \(\omega T_d = 6\pi\), the distributions become wider and the electron yield becomes much higher. This is shown in the second column of figure 1 (figures 1(b), (f), (j) and (n)). The increase in width suggests that the radius of the hypersphere (13), which delimits the classically allowed region, increased. Physically, this would correspond to a larger kinetic energy \(E_{ret}(t_1)\) of the first electron upon return. The growth in electron yield hints at an increase in the tunnelling probability for the first electron. The distributions, however, exhibit a similar qualitative behaviour to that observed for \(\omega T_d = 8\pi\), in the sense that they are asymmetric and depend on the CE phase.

A closer inspection, however, reveals a dramatic change, in the sense that the electron distributions are concentrated in an opposite momentum region, as compared to their counterparts at the delay phase of \(8\pi\). For example, if the CE phase is \(0.5\pi\), such distributions are almost entirely localized in the first quadrant of the parallel momentum plane for \(\omega T_d = 6\pi\) (figure 1(b)), while for \(\omega T_d = 8\pi\), they occupy the negative momentum region (figure 1(a)). This sharp contrast persists for the other values of CE phases. The reason behind this shift will be addressed later.

If the delay between both pulses is decreased further, the overall asymmetry in the electron-momentum distributions starts to fade. This may be seen in the third column of figure 1 (figures 1(c), (g), (k) and (o)), for which \(\omega T_d = 4\pi\). Such distributions are only slightly asymmetric and exhibit bright spots in both the first and third quadrants of the \((p_{1\parallel}, p_{2\parallel})\) plane. This trend persists for an even shorter delay of \(\omega T_d = 2\pi\), as shown in the far right panels of figure 1 (figures 1(d), (f), (l) and (p)). In this latter case, the distributions are nearly symmetric and the information about the CE phase is almost lost.

This effect is present regardless of the type of interaction by which the second electron is dislodged. In fact, for comparison, in the lowest panels of figure 1 (figures 1(q), (r), (s) and (t)), we depict electron-momentum distributions computed for the same parameters as in figures 1(a)-(d), employing, however, the form factor (17). In this case, the distributions are no longer isotropic, but exhibit a butterfly-shaped structure, which is the fingerprint of the Coulomb interaction [13]. Their behaviour with regard to the time-delay variation and its initial asymmetry for the carrier-envelope phase of \(\phi = 0.5\pi\) are very similar to those observed in the four upper panels of figure 1. In fact, the distribution in figure 1(q), similarly to that shown in figure 1(a), is asymmetric and fully concentrated in the third quadrant of the \((p_{1\parallel}, p_{2\parallel})\) plane. For a shorter delay \(\omega T_d = 6\pi\) (figure 1(r)), it has also shifted mainly to the first quadrant of such a plane, in agreement with the behaviour seen in figure 1(b). Finally, as the delay \(\omega T_d\) decreases, the distributions also become increasingly symmetric, as observed in the contact-interaction scenario.

The above-mentioned fading in the asymmetry is rather counterintuitive, as the electron-momentum distributions for short time delays \(\omega T_d\) resemble more those obtained for a linearly monochromatic driving field than what is expected for a combination of two few-cycle pulses. In the former case, the distributions are totally symmetric upon \((p_{1\parallel}, p_{2\parallel}) \rightarrow (-p_{1\parallel}, -p_{2\parallel})\). In our previous work [10], we observe that this symmetry holds in practice if the pulse is longer than ten cycles. However, the longest quasi-linearly polarized pulse considered here has no more than four cycles, as the gate width must be less than the laser-pulse duration. Therefore, intuitively, one would anticipate asymmetric distributions. In the following, we will give an explanation for such a surprising result based on lateral electron dynamics.

For this purpose, we will have a closer look at how the ellipticity of the combined laser field varies, with respect to the delay phase \(\omega T_d\). These results are displayed in figure 2, and show that the delay phase \(\omega T_d\) has a strong influence on the ellipticity of the very centre portion of the combined laser field. A longer delay phase corresponds to a sharper slope. When \(\omega T_d = 8\pi\), the ellipticity drops very steeply to zero around \(\omega_0 = 0\) and changes back soon after that. Hence, for longer time delays \(T_d\), the start times \(t_0\) of the first electron are restricted to a very narrow interval and the gate behaves better. This sheds some light on why the results in the first column

![Figure 2](image)
Figure 3. Ionization and recollision rates together with the final electron momentum along \( p_{\parallel} \) as functions of the initial phase \( \omega t_0 \), for a pulse described by equations (3) and (4), with the CE phase \( \phi = 0.5\pi \). Panel (i) exhibits the calculated ADK ionization rate. Panel (ii) displays the final recollision rate, calculated with the ADK rate and lateral velocity distribution rate. Panel (iii) gives the final momentum \( p_{\parallel} \). The tags (a), (b), (c) and (d) correspond to the delays \( \omega T_d = 8\pi, 6\pi, 4\pi \) and \( 2\pi \), respectively. The numbers \((j, \nu)\) in panel (iii) indicate a pair of electron orbits. The remaining parameters are the same as in figure 1.

of figure 1, computed for \( 8\pi \) delay phase, are similar to those obtained with linearly polarized driving fields. In contrast, we have verified that variations in the CE phase do not change the ellipticity of the combined laser field.

In figure 3, we analyse the behaviour of the ionization rate for the first electron and the phase-space contributions with respect to delay phase \( \omega T_d \). We consider parallel momenta along the diagonal, i.e., \( p_{\parallel 1} = p_{\parallel 2} = p_{\parallel} \), and impose that the perpendicular momentum components vanish. The classically allowed momentum region is most extensive for vanishing transverse momenta. Hence, we expect that the figure will provide a rough upper bound for it. We also fix the CE phase at \( 0.5\pi \). A similar analysis has been performed in [10] for linearly polarized few-cycle pulses.

In the two upper panels (figures 3(i) and (ii), respectively), we display the calculated ADK ionization rate in the central portion of the combined pulse and the final recollision rate for the first electron. The tags (a), (b), (c) and (d) in the figure follow those in figure 1, i.e., from (a) to (d), the delay phase is varied from \( 8\pi \) to \( 2\pi \). The recollision rate is weighted according to equation (9) and decided by three factors, i.e., the tunnelling ionization rate, the wave packet spread time and the quantum-mechanical transverse velocity distribution weight. Since the lateral electric field of the combined laser pulse will shift the electron wave packet laterally, trajectories with zero initial lateral velocity, which may return to the core in a linearly polarized laser, will miss the core. Only those with a certain initial lateral velocity, which may compensate for the displacement induced by the lateral electric field, can return to the core and contribute to the NSDI yield. The effect of the transverse velocity has been demonstrated in HHG by a recent experiment, where an elliptical laser pulse was employed [20].

From equation (8), one can see that, as compared to the trajectories with a vanishing initial lateral velocity, those with a nonvanishing initial lateral velocity have a lower transverse velocity distribution weight, which in turn reduces the final recollision rate. Therefore, the recollision rate will be corrected according to the transverse electron dynamics. Comparing figures 3(i) and (ii), one also finds that the highest recollision rate appears slightly later than the peak of the ADK ionization rate. This is a consequence of the fact that the trajectories starting near the peak laser electric field will propagate with longer times and be affected more by the wave-packet spreading effect. This effect results in the drop of the recollision rate according to a cubic power law with the evolution time \( t_1 - t_0 \).

The parallel momenta \( p_{\parallel} \) in figure 3(iii) provide an approximate estimate for the region in momentum space for which electron-impact ionization is classically allowed. The region delimited by such momenta gives a very good idea of the role of phase-space effects: the larger this region is, the more important they are. A small region, on the other hand, means a small radius for the hypersphere in equation (16), which, physically, indicates that the second electron may only be dislodged in a small region in momentum space. In figure 3(ii), one may identify a few sets of orbits, which lead to parallel momenta either in the positive or in the negative region. Starting from the left, these orbits are denoted by \((1, 2), (3, 4), (5, 6)\) and \((7, 8)\).

The momentum region to which they contribute depends on the time delay \( \omega T_d \). If this delay is an even multiple of the field cycle \( 2\pi \), i.e., for \( \omega T_d = 4\pi \) and \( \omega T_d = 8\pi \), there exist at most two sets of orbits, \((1, 2)\) and \((5, 6)\), which yield contributions in the first quadrant of \((p_{\parallel 1}, p_{\parallel 2})\). These orbits start within \(-2\pi < \omega t_0 < -\pi \) and \(0 < \omega t_0 < \pi \), respectively. The remaining sets of orbits, starting at \(0 < \omega t_0 < \pi /2\) and \(3\pi /2 < \omega t_0 < 2\pi \), lead to negative parallel momenta. If \( \omega T_d \) is an odd multiple of the field cycle, i.e., for \( \omega T_d = 6\pi \) and \( \omega T_d = 2\pi \), the above-mentioned situation is reversed.

For linearly polarized driving fields, the ADK ionization rate and phase-space effects suffice, in order to determine whether a set of orbits contributes significantly to the electron-momentum distributions. A large rate indicates that the first electron will tunnel with a significant probability per unit time. Furthermore, if this electron returns with sufficient energy to release the second electron over a significant region of momentum space, one expects that the contributions from a specific set of orbits will be prominent [10, 11]. For a polarization-gated pulse, however, we have to take into account the lateral electron dynamics. This information is embedded in the recollision rate depicted in figure 3(ii).

We will now analyse the interplay of the above-mentioned issues in the electron-momentum distributions. For a delay \( \omega T_d = 8\pi \), there is mainly a single set \((3, 4)\) of trajectories for which electron-impact ionization is classically allowed. This set corresponds to \( \omega t_0 \sim \frac{-\pi}{2} \) and leads to contributions in the negative momentum region. The contribution from another set \((5, 6)\) of orbits may be ignored due to its small classical allowed region and low recollision rate. For this reason, the distributions are highly asymmetric and concentrated in the third quadrant of the parallel momentum plane \((p_{\parallel 1}, p_{\parallel 2})\), in agreement with figure 1(a). This very restricted range in the
classically allowed region is possibly due to the fact that the ellipticity of the driving field changes very fast. Hence, the first electron only returns to its parent ion within a very narrow temporal region. The tunnelling rate is also weak in this case, as it is taken only in the central part of the combined pulse and there is very little overlap between each circularly polarized pulse for this specific delay.

As the delay phase decreases to $\omega T_d = 6\pi$, the situation becomes different. In this case, the orbits $(3, 4)$ lead to $p_1 > 0$. Hence, the distributions are concentrated in the positive parallel momentum region. Apart from that, there is also a further set $(5, 6)$ of orbits, whose start times lie near $\omega t_0 \sim -\frac{\pi}{2}$. The contributions of this latter pair are weaker, and localized in the third quadrant of the plane $(p_{1\parallel}, p_{2\parallel})$. Finally, as an overall feature, there is an increase in the ADK rate and also in the recollision rate for the first electron. This is due to an increase in the overlap between the two pulses $E_r$ and $E_l$, and leads to brighter distributions. All the above-stated features can be observed in figure 1(b).

A further reduction in the delay phase to $\omega T_d = 4\pi$ leads to an additional set $(7, 8)$ of orbits for which the first electron may return and release the second electron. This set starts near $\omega t_0 \sim -3\pi/2$ and leads to contributions in the first quadrant of the plane $(p_{1\parallel}, p_{2\parallel})$. This will add up to the contributions from the orbits $(5, 6)$. Hence, overall, there will be two sets of orbits yielding momenta in such a region.

The ionization and recollision rates, however, are larger for the orbits $(3, 4)$ starting at $\omega t_0 \sim -\pi/2$. Therefore, the distributions are slightly brighter in the negative momentum region. Finally, for $\omega T_d = 2\pi$, there are four sets of orbits contributing to the electron-momentum distributions, and the distributions are approximately symmetric.

In figure 4, we display the initial lateral velocity for the rescattering orbits (upper panel) and the lateral laser electric field (lower panel) as functions of the initial electric field phase $\omega t_0$. Classically, for a linearly polarized driving electric field, an electron with a large lateral initial velocity will miss the core. For an elliptically polarized field, however, the lateral electric field will also slightly change the electron’s orbit in the transverse direction.

If both effects are combined, a gate can be formed to choose the orbit with a certain initial lateral velocity to come back with highest probability [20]. A larger lateral velocity corresponds to a stronger lateral laser electric field and, according to equation (7), to a lower ionization rate. In figure 4, the orbits starting near $\omega t_0 \sim -\frac{\pi}{2}$ meet the smallest lateral laser electric field because their tunnelling phases are those nearest to the centre of the gate. Hence, these orbits exhibit the largest recollision probability. This explains why we observe a favoured NSDI rate in this region in figure 3.

It is also noteworthy that this specific set of orbits exhibits lateral velocities close to zero, whereas the lateral velocities of the remaining sets vary considerably with the time delay. In general, as the delay phase $\omega T_d$ decreases, the lateral velocities diminish as well. This causes an overall increase in the tunnelling and recollision rates, in agreement with figure 3.

Apart from that, specifically for the time delay $\omega T_d = 2\pi$, there is an overall decrease in the lateral velocities for the orbits starting near $\omega t_0 \sim -\pi$ and $\omega t_0 \sim \pi$. Both sets of orbits lead to positive final momenta $p_{n\parallel}(n = 1, 2)$. Thus, we will expect an increase in brightness in the first quadrant of the $(p_{1\parallel}, p_{2\parallel})$ plane. As a direct consequence, the distributions are nearly symmetric. The set of orbits $(7, 8)$, starting near $\omega t_0 = 1.5\pi$, exhibits very large transverse velocities and therefore does not contribute significantly to the yield.

Still, one question that remains is how to understand the dramatic change in the electron-momentum distributions between the delay phase of $8\pi$ and $6\pi$, as shown in figure 1. For this purpose, we further conduct the calculations for the CE phase $\phi = 0.5\pi$ and several delay phases between $8\pi$ and $6\pi$, i.e., $\omega T_d = 7.5\pi, 7\pi$ and $6.5\pi$. The results are displayed in figure 5. In addition to the momentum components $(p_{1\parallel}, p_{2\parallel})$ in the $x$-direction, we also consider the momentum components $(p_{1\perp}, p_{2\perp})$. The distributions as functions of the former or latter momentum components are displayed in the upper and lower panels of the figure, respectively. Therefore, for the sake of clarity, we no longer employ the notation $p_{n\parallel}$.

It is found that, actually, the electron-momentum distributions evolve gradually from the third quadrant to the first quadrant, with the decrease of the delay phase between $8\pi$ and $6\pi$. Interestingly, at a delay phase of $7\pi$, the electron distribution is almost centred at the origin (figure 5(c)). This is quite different from the case with a linearly polarized few-cycle pulse, for which the centre of the electron-momentum distribution is always at nonvanishing momenta and the distributions just ‘jump’ from one quadrant to another. This is due to the fact that, in the linearly-polarized few-cycle case, the ‘jump’ is caused by a shift in the dominant set of orbits, while in the present situation, the dominant set of trajectories remains the same. The changes occur in the momentum components in the $x$-direction.

This modification in the most probable momenta is due to the fact that the polarization gate changes direction within this phase-delay interval. In fact, as the $x$-momentum components $p_{n\parallel} (n = 1, 2)$ decrease, there is a corresponding increase in
the two combining field components in the \( x \)-direction are totally out of phase. While for \( \omega T_d = 8\pi \) and \( 6\pi \), the fields have maximal amplitudes but opposite directions due to their relative phase shift of \( \pi \).

As discussed in section 2, the centre of the hypersphere which delimits the momentum distributions in momentum space is determined by the vector potential-component \( A_z(t) \) at the instant when the electron recollides. For \( \omega T_d = 7\pi \), the amount of momentum the electron acquired from the combined field is thus very small in the \( x \)-direction. Hence, the classically allowed region is approximately centred at momentum \( p_x = 0 \), as shown in figure 6(iii). For \( \omega T_d = 8\pi \) and \( 6\pi \), the relative shift of \( 2\pi \) in the delay phase results in the inversion of the \( x \)-component of the combined field at the very centre of the polarization gate. This gives rise to electron emission in the opposite direction, i.e., the electron-momentum distribution shifts from the third to the first quadrant.

4. Conclusions

In this work, we have studied laser-induced nonsequential double ionization (NSDI) with a polarization-gated driving pulse. The specific pulse employed here consisted of two few-cycle pulses with opposite circular polarizations and a time delay \( T_d \) (for details we refer to [5]). The ellipticity of the combined field is time dependent and vanishes for \( t_0 = 0 \). The larger the time delay between both pulses, the steeper the time dependence in the polarization. We performed such studies within a classical framework, extending the model in [10, 11, 13] to elliptically polarized fields.

We found that the electron-momentum distributions, as functions of the electron components parallel to the laser-field polarization at the centre of the gate, are very much dependent on the delay between the two few-cycle pulses \( \vec{E}_c \) and \( \vec{E}_l \) composing the polarization-gated pulse. For long delays that are chosen as multiple times of the pulse duration, the distributions are asymmetric and dependent on the CE phase of such pulses. This is similar to the behaviour observed for a single linearly polarized few-cycle driving pulse [10, 11].
As the time delay $T_d$ decreases, this asymmetry fades. We could explain this behaviour in terms of trajectories. Below, we will summarize the main aspects of this explanation and also draw a parallel between the present situation and the previously studied case of linearly polarized few-cycle pulses [10, 11].

For linearly polarized few-cycle pulses, the momentum region in which the electron momentum distributions will be concentrated depends on the quasi-static tunnelling rate for the first electron and on the momentum region for which NSDI is classically allowed. A large rate and a large momentum region imply that the contributions from a specific set of trajectories to the distributions will be prominent. For very short pulses, there is in general a single set of trajectories that best fulfils such conditions. This set will change with the CE phase. As the pulse length increases, more and more sets of trajectories will lead to prominent contributions, and the asymmetry will fade.

For polarization-gated pulses, the most relevant set of orbits will be that closest to the polarization gate, i.e., to the quasi linearly polarized region of the pulse. For this specific pair, the first electron will tunnel more efficiently than for the remaining sets of orbits. This is due to the fact that the electron velocity components perpendicular to the main polarization axis of the laser field are smallest for the dominant set of orbits.

Apart from that, the first electron will return more easily to the parent ion if it is released near the polarization gate. This will happen for two main reasons. First, the small lateral velocity components will not lead to a significant initial motion in the transverse direction. Second, the electron will propagate in the continuum in a time interval close to the gate. Hence, it will be accelerated in the transverse direction to a lesser extent than for the remaining sets of orbits. This will guarantee an efficient return to the ion.

For an efficient polarization gate, the changes in the driving-field ellipticity within the pulse length are very steep, so that only the orbits close to the gate will contribute significantly to the distributions. In general, this will lead to highly asymmetric electron momentum distributions, concentrated in the momentum region determined by the dominant orbits. This is a similar behaviour to that encountered for linearly polarized driving fields, in the sense that, in general, only one set of trajectories dominates the yield.

If the gate, however, is less efficient, the contributions from the other sets of orbits will be increasingly important. Ultimately, this will lead to nearly symmetric electron-momentum distributions. For the specific driving pulse in this work, this effect has been observed by decreasing the delay $T_d$ between the pulses.

It is also noteworthy that the effects reported in this work are present regardless of whether the second electron is dislodged by a contact-type or a Coulomb interaction. This is due to the fact that the asymmetry of the distributions for different CE phases, and the fact that it gradually vanishes for smaller time delays are phase-space related. Hence, even if there is some momentum bias due to the fact that the form factor $V_{p,ij,k}$ is no longer constant, the classically allowed region for electron-impact ionization is altered in the very same way as for the contact-type interaction as the CE phase, polarization and pulse lengths are changed.

A similar situation is encountered for NSDI with linearly polarized few-cycle pulses. In the linearly polarized case, the asymmetry in the electron-momentum distribution is caused by the fact that whole momentum regions for which electron-impact ionization is classically allowed appear or collapse, as the CE phase is varied. This effect is so extreme that a simple model based on a contact-type interaction [10] has enough predictive power to foresee effects measured experimentally [9].

In general, the efficient use of polarization gates could tremendously increase the degree of attainable control of strong field ionization dynamics, compared to the commonly used single linearly polarized few-cycle pulse. For the latter, only the scalar properties of laser pulses are used, while a polarization gate can, in principle, as demonstrated in this work, make full use of the flexibility of the vectorial laser field, e.g., the polarization state, as an additional knob to steer the NSDI dynamics within sub cycle time resolution.

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