Controlling sub-cycle electron dynamics with plasmonic bichromatic counter-rotating circularly polarised fields

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The use of bi-chromatic counter-rotating laser field is known to generate high-order harmonics with non-zero ellipticity. By combining such a laser pulse with a plasmonic field, we propose a way to influence the sub-cycle dynamics of the high-harmonic generation process. Using a numerical solution of the time-dependent Schrödinger equation combined with classical trajectory Monte Carlo simulations, we show that the change of the direction and the strength of the plasmonic field selectively enhances or suppresses certain recombining electron trajectories. This in turn modifies the ellipticity of the emitted attosecond pulses.

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Control over the polarisation of attosecond pulses in extreme ultraviolet (XUV) and soft x-ray radiation is paramount to probing chiral-sensitive light-matter interactions such as x-ray magnetic circular dichroism [1, 2], discrete molecular symmetries [3, 4], magnetisation and spin dynamics [5, 6], and recognising chirality in molecules via photoelectron circular dichroism [7, 8] at their intrinsic timescales. Following the proposal of Becker and co-workers [9, 10], a series of experiments have been carried out to generate high harmonics with controlled polarisation [1, 2, 11, 12]. A combination of two counter-rotating circularly polarised laser pulses having fundamental ($\omega$) and its second harmonic (2$\omega$) frequency is employed to generate circularly (or elliptically) polarised high-harmonics. The resultant electric field of $\omega$-2$\omega$ combination exhibits trefoil symmetry and yields three radiation bursts per cycle of the fundamental field [13, 14, 15, 16, 17]. The resulting high-harmonic spectrum contains doublets of circularly polarised harmonics with alternating helicity. The 3n+1 and 3n+2 harmonics follow the polarisation of $\omega$- and 2$\omega$-fields, respectively, whereas 3n harmonics are parity forbidden [18, 19, 20, 21].

However, in High-Harmonic Generation (HHG) the control over the polarisation of the harmonics does not immediately entail the same control over the polarisation of underlying attosecond pulses. Therefore, proposals have been put forward in recent years to achieve the subcycle emission control by either modifying the underlying medium, (e.g., by choosing a suitable initial state of atoms [21–24, 25], using different molecular systems [26, 27], or by modifying the laser driving schemes, (e.g., by using non-collinear bichromatic counter-rotating circularly polarised laser pulses [28, 29], introduction of seed XUV pulse with suitable polarisation [30], tuning the ratio of the ellipticity and/or of the intensity of the bichromatic counter-rotating driving fields [19, 20, 31], or by changing the time-delay between two driving pulses [19, 32]).

In this work, we demonstrate the control over sub-cycle electron dynamics during HHG by combining bichromatic counter-rotating circularly polarised driving fields (BiCRCP) with plasmonic field enhancement. The direction and strength of the plasmonic field will provide the desired control over sub-cycle electron dynamics, which in turn will allow us to exert influence over the polarisation of attosecond pulses.

Since the experiment of Kim et al. [33], a number of works have investigated the plasmonic-field assisted HHG using structured nano-objects [33, 34]. Moreover, different kinds of nanostructures such as metal nanotips [35, 36], metallic waveguides [37], nanoparticles [38], and plasmonic antennas [39], have been explored, while many more works tried to explain HHG mediated by a plasmonic field using numerical and theoretical methods [40, 41]. Despite the abundance of research, most of the works on plasmonic-field assisted-HHG are investigating linearly polarised driving fields. In this regard, its not apparent how the high-harmonic spectrum will behave when the linearly polarised pulse in the presence of plasmonic field is replaced by BiCRCP fields. Will the pattern of circularly polarised doublets with alternating helicity be preserved throughout the spectrum? Will the increase in the energy cutoff be modified?

In order to study these questions, the time-dependent Schrödinger equation (TDSE) is solved in two-dimensions as discussed in Ref. [21]. Atomic units are used throughout in the present work. The following model potential

$$V(r) = -\frac{1 + 9 e^{-r^2}}{\sqrt{r^2 + a}},$$

with the parameter $a = 2.88172$ is used to describe a 2D model neon atom with the ionisation potential of the initial p-orbital equal to $I_P = 0.793$ a.u. (21.6 eV) [68]. The driving infrared (IR) field has the following form:

$$E_{\text{IR}}(t) = E_0 g(t) \left[ E_x(t) \hat{x} + E_y(t) \hat{y} \right].$$

Here, the magnitude of the field $E_0$ is 0.05 a.u., and $g(t)$ is a trapezoidal envelope with five-cycle plateau and two-
cycle rising and falling edges. $E_x(t) = \cos(\omega t) + \cos(2\omega t)$ and $E_y(t) = \sin(\omega t) - \sin(2\omega t)$ are the $x$- and $y$- components of the IR field, respectively, where $\omega = 0.05$ a.u. is the fundamental frequency.

The plasmonic field enhancement is described by including a space-dependent field component

$$E^{pl}(x, y, t) = E_0 g(t) [\beta_x h(x) E_x(t) \hat{x} + \beta_y h(y) E_y(t) \hat{y}],$$

(3)

to the total electric field $E^{total} = E^{pl}(t) + E^{ff}(t)$. Parameters $\beta_x$ and $\beta_y$ characterises the strengths of plasmonic field along the $x$- and $y$-directions, respectively, and has the dimensions of the inverse length. The spatial inhomogeneity $h$ is typically approximated to linear order, i.e., $h(x) = x$ and $h(y) = y$, which is the chosen form in our model, too.

Figure 1(a) shows the reference harmonic spectrum obtained for the homogeneous case with $E^{ff} = 0$. The spectrum consists of $3n + 1$ and $3n + 2$ harmonics exhibiting right (red) and left-handed (blue) circular polarisations, respectively, and the $3n$ harmonics are absent. This was studied in detail in previous works [5, 19, 21, 23, 26]. The reference spectrum also shows a clear energy cutoff at around $\approx 50^{th}$ harmonic order, which is consistent with the cutoff law for BiCRCP fields of equal intensity [45]. Elliptically polarised attosecond pulses are expected since the spectral intensities of harmonics with opposite helicity are different [21].

Figures 1(b) and (c) present plasmonic field assisted harmonic spectra with $\beta_x = 0.01$ a.u. and $\beta_y = 0.01$ a.u., respectively. The spectra are drastically different in comparison to the reference case of Fig. 1(a). First, there is a significant increase in the energy cutoff, which is a general feature of plasmonically-enhanced HHG [47, 50, 62, 64]. Second, harmonics of order $3n$ appear in the spectra for higher orders, which is also expected, since the spatially-dependent plasmonic field $E^{pl}$ breaks the trefoil symmetry of the total field $E^{total}$. Furthermore, above $35^{th}$ order the right- and left- circularly polarized harmonics overlap with approximately equal intensities and have no apparent selection rules. This is a strong indication that the harmonics above $35^{th}$ order originate from linearly or low-elliptically polarised attosecond pulses generated once per laser cycle. Note, that the differences in the spectra are sensitive to the direction of the plasmonic field since the spectra in Figs. 1(b) and (c) are different.

To connect the changes of the harmonic spectra with the underlying sub-cycle electron dynamics, we obtain time-frequency maps by performing a Gabor transform of the time-dependent dipole

$$\hat{d}_{x,y}(\Omega, t) = \frac{1}{2\pi} \int dt \, d_{x,y}(t) \, e^{-i\Omega t} e^{-\frac{(t-\tau_0)^2}{(2\sigma^2)}}$$

(4)

with $\sigma = 1/(3\omega)$. The time-frequency map of the reference spectrum shows three bursts of radiation of equal strengths produced during every cycle $T \approx 2.6$ fs of the fundamental field (see Fig. 1(d)). It can be understood by the Lissajous figure of the total field, which shows that the total electric field reaches its maximum three times during a single cycle of the fundamental field (see inset in Fig. 1(g)).

To further interpret the continuum electron dynamics and underlying HHG process, we use classical trajectory Monte Carlo (CTMC) simulations. Details of the CTMC calculation can be found in [45] and references therein. In addition to an ADK initial transverse momentum distribution [66, 67], a Gaussian distribution of initial longitudinal momentum was allowed [68, 69].

The return-times and -energies of the ionized electrons in the reference driving field obtained from the CTMC calculations are shown in Fig. 1(g). The return condition for each trajectory was a shorter distance to the ion than the individual exit radius [65, 70]. The CTMC trajectories are grouped according to the ionisation events occurring during one of the three lobes of the driving field, indicated by different colors in Fig. 1(g). Comparing Figs. 1(d) and (g) we see that the CTMC calculations reproduce the Gabor analysis well. Furthermore, it is made clear that the main features in the reference spectra are produced by the short electron trajectories, i.e., the electrons that return during the next lobe of the field after their ionization. This is in good agreement with trajectory-based studies of HHG in bi-chromatic counter-rotating field [71].

As we have seen in Figs. 1(a-b), the plasmonic field tends to break the symmetry of the harmonic spectra. In contrast, the time-frequency maps in Figs. 1(b-c) reveal a very regular sub-cycle dynamics. The effect of the (symmetry-breaking) plasmonic field is to enhance (or suppress) the energy-cutoff of attosecond bursts of radiation produced during each lobe of the driving laser field. When the plasmonic field is along the $x$-direction (parallel to one of the field maxima directions) in Figs. 1(b,c) and (h), those short trajectories which are ionised during a lobe immediately before the laser field aligns with the plasmonic field have enhanced kinetic energy. The picture changes completely when the plasmonic field is oriented along $y$-direction (30° from the nearest field maximum) in Figs. 1(c),(f) and (i). Here, the short trajectories ionised during the second lobe of the field return with reduced energy, while short trajectories ionised during the first lobe and long trajectories from both others contribute to the enhanced harmonics.

The enhancement of attosecond radiation bursts becomes clear, when looking at the probability density maps of returning trajectories in Fig. 2. They are obtained by CTMC simulation, for ionisation during each lobe of the total field and for the plasmonic field applied along $x$-direction ($\beta_x = 0.01$ a.u.). If the electron is ionized during the lobes 1 or 2, the excursion of its returning trajectory can be much further and these electrons can recombine during the next several lobes after ionization.
[see Figs. 2(a) and (b)]. Contrarily, only short trajectories return when an electron is ionised during lobe 3 and recombines during lobe 1 of the next cycle [see Fig. 2(c)].

Trajectories returning towards the ion from the positive $x$-axis are those which yield harmonics with enhanced cut-off energy [compare Fig. 2 and Fig. 1(a)]. In this case, both the direction of the applied plasmonic field and the direction of the driving field coincides with the direction of returning photoelectron such that the electron feels the maximum acceleration upon return and accumulates more kinetic energy. Similar reasoning can be used if we apply the plasmonic field along $y$-direction ($\beta_y = 0.01$ a.u.). In this case, higher energy photons are produced when photoelectrons recombine during lobe 2 of the field, see again Fig. 1(i). By reversing the helicity of the laser field, i.e., choosing negative helicity for the $\omega$-field and positive helicity for the $2\omega$-field, while keeping $\beta_y = 0.01$ a.u., the short trajectories returning at approximately 12 fs will be enhanced because they are ionised when the field points mostly along negative $y$-direction and return from positive $y$-direction during the next lobe (not shown here).

So far we have discussed only the results for a plasmonic field with strength $\beta_{x,y} = 0.01$ a.u., for which the polarisation and position of the harmonics below the cut-off energy remain largely unaffected. As the strength of the plasmonic field is increased to $\beta_{x,y} = 0.02$ and 0.05 a.u., the below-cutoff harmonic spectrum undergoes a substantial change with the appearance of $3n$ harmonics, and right- and left-handed harmonics overlapping with equal strength. This modification is driven by the change of polarisation of the underlying attosecond pulses. We demonstrate this change by plotting, in Fig. 3, the total electric field and its $x$- and $y$-components for a window between the $18^{th}$ – $24^{th}$ harmonics and for different plasmonic field strengths.

For the homogeneous case in Fig. 3(a), the attosecond pulses have elliptical polarization, where the major polarization axis is rotated by 120° for each pulse. This is a direct consequence of unequal intensities of the $3n+1$ and $3n+2$ harmonics [19, 21, 23, 33]. Indeed, if
FIG. 2: Classical electron trajectories, which are returning back to the parent ion, ionised during three different lobes of the field. The plasmonic field is employed along $x$-direction with $\beta_x = 0.01$ a.u. Some exemplary trajectories are highlighted by black arrows. The insets indicate the lobe during which the ionization takes place and the rotation direction of the laser electric field vector.

Beyond the harmonic cut-off energy, the emitted radiation has linear polarization with a direction that coincides with that of the applied plasmonic field. Consequently, all the harmonics including $3n$-harmonics have comparable intensities. For example, the electric field obtained by filtering out $60^{th} - 66^{th}$ harmonics has a linear polarization along $x$- ($y$-) direction when $\beta_x \neq 0.00$ ($\beta_y \neq 0.00$). The same is true for the electric field generated by $78^{th} - 84^{th}$ harmonics.

This observation corresponds to what we have already seen from the CTMC analysis. The HHG spectrum above the reference cut-off energy stems from the photoelectrons that return to the parent ion from the direction of the plasmonic field and therefore, are additionally accelerated to higher kinetic energy. However, since the additional acceleration occurs only along the plasmonic field direction, the high-energy radiation is linearly polarized and is emitted only once per laser cycle. Any photoelectron trajectories returning and recombining from a different angle will only contribute to the lower energy part of the HHG spectrum.

In summary, while the HHG process driven by Bi-CRCP laser field follows the three-fold symmetry pattern, adding a local plasmonic field along a particular spatial dimension modifies the sub-cycle electron dynamics in non-trivial ways: enhancing some long or short photoelectron trajectories while suppressing some others. Simply by changing the direction of plasmonic field enhancement, rich yet predictable control of electron trajectories is possible, providing as yet unexplored possibilities for sub-cycle control of the HHG process. Furthermore, the modification of the sub-cycle electron dynamics also
leads to subtle changes in the polarization of the emitted high-harmonic radiation. For instance, by filtering appropriate harmonic orders, an attosecond pulse train consisting of nearly-circularly, elliptically or linearly polarized pulses is achieved. Alternatively, by changing the plasmonic field intensity, predefined major axis direction is enforced for all elliptically polarized pulses in an attosecond pulse train.

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