The influence of the driving-bicircular-field component intensities on the helicities of emitted high-order harmonics

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Abstract. High-order harmonics generated by a linearly polarized laser field are also linearly polarized. Having in mind that for various application, such as the exploration of magnetic materials, chiral molecules etc., we need circularly polarized high harmonics which serve as coherent soft x-rays, we explore high-order harmonic generation by the so-called bicircular laser field. This field consists of two coplanar counter-rotating circularly polarized fields of different frequencies equal to integer multiples of a fundamental frequency \( \omega \). High harmonics generated by such field are circularly polarized with helicity alternating between \(+1\) and \(-1\). Combining a group of such harmonics, instead of obtaining a circularly polarized attosecond pulse train, one obtains a pulse with unusual polarization properties. But, if the harmonics of particular helicity are stronger, i.e., if we have helicity asymmetry in a high-harmonic energy interval, then it is possible to generate an elliptical or even circular pulse train. We theoretically investigated a wide range of bicircular field-component intensities \( I_1 \) and \( I_2 \) and found regions where both the harmonic intensity is high and the helicity asymmetry is large. Particular attention is devoted to the \( \omega - 2\omega \) and \( \omega - 3\omega \) bicircular fields and atoms having the \( s \) and \( p \) ground states. In our calculations we use strong-field approximation and quantum-orbit theory. We show that, even in the extreme case of \( I_2 = 8I_1 \), for an \( \omega - 3\omega \) bicircular field, high-order harmonic generation is more efficient than in the \( I_2 = I_1 \) case. The obtained results are explained analyzing the relevant electron trajectories and velocities, which follow from the quantum-orbit theory. For the atoms having \( p \) ground state the helicity asymmetry parameter is large for a wide range of high-harmonic photon energies, while for the atoms having \( s \) ground state the helicity asymmetry parameter can be large only for low harmonics. We confirm this by averaging the obtained results over the intensity distribution in the laser focus.

1. Introduction
High-order harmonic generation (HHG) is a strong-laser-field-induced process in which the energy absorbed from the laser field is emitted in the form of a high-energy photon. This process can be explained using the semiclassical three-step model [1]. In the first step the electron is liberated at time \( t_0 \) in atomic tunnel-ionization process induced by a strong linearly polarized laser field. The liberated electron, driven by the Lorentz force moves away from the parent ion and may return to it after the field changes the sign. This is the second step. Finally,
in the third step, the electron, returned to the core at the time $t_1$, recombines to the atomic ground state and the atomic ionization potential energy $I_p$ plus the returned electron kinetic energy is released in the form of a high-energy photon. The probability of the HHG process is approximately the same for all photons with energies larger than $I_p$ so that the corresponding HHG spectrum forms a plateau. This plateau is followed by a cutoff.

The laser field in the above-described three-step model is linearly polarized and so is the emitted high-harmonic field. It is well known that HHG is not possible with circularly polarized laser field since the liberated electron, driven by such field, cannot return to the parent ion to recombine. However, it is possible to generate high-order harmonic using the so-called bicircular laser field. This field consists of two coplanar counter-rotating circularly polarized laser fields having different frequencies. That HHG by bicircular field is very efficient was confirmed experimentally in 1995 [3] (see also theoretical papers [4, 5]). In 2000 HHG by bicircular field was explained in the spirit of the above-described three-step model using the quantum-orbit theory [6]. Two-dimensional electron trajectories which return to the parent ion were identified. Furthermore, it was found that the emitted high-order harmonics are circularly polarized with alternating ellipticities equal to ±1. This was confirmed experimentally in 2014 [7]. For application it is important to generate circularly polarized high-order harmonics which can serve as a source of soft x-ray photons. Such photons have application for analysis of various chirality sensitive processes in organic molecules [8, 9], magnetic materials [10, 11] etc.

Combining a group of circularly polarized high-order harmonics having ellipticities which alternate between +1 and −1, instead of obtaining a circularly polarized pulse, we obtain a pulse with unusual polarization properties. This was first shown in [12] where, for bicircular field with frequencies $\omega$ and $2\omega$, a star-like structure with three linearly polarized pulses rotated by $120^\circ$ was obtained. This theoretical prediction was recently confirmed experimentally [13]. It was suggested in 2001 [14] that circularly polarized attosecond pulse trains can be generated if the harmonics having helicity +1 are stronger than that of helicity −1 (and vice versa), i.e. if we, by some means, achieve helicity asymmetry in a high-harmonic photon energy interval. In [14] such asymmetry was noticed for He atoms, having $s$ ground state, in the case when the intensity of the $2\omega$ bicircular field component was two times higher than that of the $\omega$ component. Later on, in 2015, it was found that a strong helicity asymmetry for much higher photon energies exists for HHG by inert gases having the $p$ ground state [15, 16, 17].

In the present work we analyze the influence of the driving-bicircular-field component intensities on the helicities of emitted high-order harmonics. After a brief presentation of the used theory, we show (and illustrate by an example) that an efficient HHG by bicircular field is not limited only to the field with component frequencies $\omega$ and $2\omega$ and with comparable component intensities. For explanation of the obtained results we use quantum-orbit theory. Next, we present results for harmonic intensity and helicity asymmetry parameter, averaged over the intensity distribution in the laser focus, for a wide range of the ratios of the intensities of the bicircular field components. At the end of this paper our conclusion is given. We use atomic system of units.

2. Theory

We consider HHG by bicircular field having component frequencies $r\omega$ and $s\omega$ ($r, s$ integers) and intensities $I_1 = E_1^2$ and $I_2 = E_2^2$. The corresponding electric-field-vector components are

$$E_x(t) = \frac{[E_1 \sin(r\omega t) + E_2 \sin(s\omega t)]}{\sqrt{2}},$$
$$E_y(t) = \frac{[-E_1 \cos(r\omega t) + E_2 \cos(s\omega t)]}{\sqrt{2}}.$$  (1)

Using the dynamical symmetry of this field the following selection rule is derived [17]

$$\varepsilon_n = \pm 1 \quad \text{for} \quad n = q(r + s) \pm r, \quad (q \text{ integer})$$  (2)
i.e., only circularly polarized high-order harmonics of order \( n = q(r + s) \pm r \) and ellipticity \( \epsilon_n = \pm 1 \) are emitted. Denoting the two-component \( T \)-matrix element for emission of the \( n \)th harmonic photon by \( T_n = T_n^x \hat{e}_x + T_n^y \hat{e}_y \), for the intensity of the \( n \)th harmonic irradiated in the direction of the \( z \) axis we obtain

\[
I_n = \frac{(n\omega)^4}{2\pi e^4} \left( |T_n^x|^2 + |T_n^y|^2 \right),
\]

where \( T = 2\pi/\omega \) is the fundamental field optical period, \( d_m(t_1) \) is the time-dependent dipole and the magnetic quantum number is \( m = 0 \) for the \( s \) atomic ground state or \( m = \pm 1 \) for the \( p \) state. The time-dependent dipole can be calculated using the strong-field approximation (SFA) by numerical integration over the electron travel time \( \tau \), as described in [17]. Alternatively, the integrals over the ionization time \( t_0 = t_1 - \tau \) and the recombination time \( t_1 \) can be solved using the saddle-point approximation (SPA). In this case the \( T \)-matrix element has the form

\[
T_n = \sum_s A_s e^{iS_s},
\]

where the sum is over \( s \equiv \{t_0s, t_1s\} \). The complex times \( t_0s \) and \( t_1s \) are solutions of the corresponding saddle-point equations, \( A_{sx} \) and \( A_{sy} \) are the amplitudes and \( S_s \) is the action. For details see [6, 18].

As we have explained in the introduction, we want to find for which bicircular-field-component-intensity ratio there is an interval of harmonic photon energies with the dominant harmonic helicity. On the other, the plateau for the SFA results is flat having an oscillatory behavior. We see that in the low-

3. Quantum-orbit analysis of the results for He, \( I_2 = 8I_1 \) and the \( \omega - 3\omega \) case

In previous papers the results for HHG by bicircular field have been calculated mainly for the \( \omega - 2\omega \) case \( (r = 1 \) and \( s = 2 \) and equal intensities of the field components. In [19] an optimization of the ratio of the component intensities was performed with the aim to obtain maximum harmonic intensity for wavelength 13 nm (in connection with the development of extreme-ultraviolet lithography). Saddle-point analysis for the \( I_2 = 2I_1 \), presented in [14], indicates that the harmonic intensity is higher in this case (in comparison with the \( I_2 = I_1 \) case). A more detailed analysis has recently been presented in [18].

In order to investigate the influence of the ratio of the bicircular field component intensities on the harmonic spectra, we will consider an extreme case of \( I_2 = 8I_1 \) and \( \omega - 3\omega \) field combination. On the first glance, since \( I_2 \gg I_1 \) and since HHG by a circular field alone is negligible, one would expect minor HHG in the considered example. However, we will show that this is not the case and we will explain why it is so. In figure 1 by dotted lines we present the SFA results for equal component intensities, separately for harmonic helicity equal to \( +1 \) and \( -1 \), denoted in the legend as SFA1+ and SFA1−, respectively. Analogous results for \( I_2 = 8I_1 \) are presented by long dashed lines and denoted by SFA8+ and SFA8−. We see that in the low-energy region the SFA1± and SFA8± results are comparable, while in the plateau region the SFA8± results are dominant. The plateau for the SFA1± results is inclined and decreasing with the increase of the harmonic order. In addition, the results practically do not depend on the harmonic helicity. On the other, the plateau for the SFA8± results is flat having an oscillatory
structure and the cutoff near $n = 200$. In order to explain this structure, we calculated partial harmonic spectra using the saddle-point approximation and two pairs of the shortest quantum orbits. Each pair is characterized by its own cutoff after which the contribution of the solution presented by the dashed line should be neglected. The contribution of the orbits SPA1 has a cutoff near $n = 150$ and the corresponding harmonic intensity is slightly higher than that of the orbits SPA2 which has a higher cutoff ($n = 200$). The interference of the SPA1 and SPA2 contributions is responsible for the mentioned oscillatory structure. Due to these oscillations it is difficult to estimate the helicity asymmetry parameter. One can see that for low harmonics this asymmetry is substantial. However, for below-threshold harmonics ($n\omega < I_p$) the saddle-point approximation is questionable. In addition, helicity asymmetry for low harmonics is less important for applications. One can also notice a region near $n = 37$ where the harmonics having helicity $+1$ are stronger than that having helicity $-1$. These are low harmonics, but above the mentioned threshold. We have checked that the asymmetry in this region appear also for $\omega - 2\omega$ bicircular field and various ratios of the component intensities (compare figure 3).

In order to further explain the results of figure 1, in figure 2 we present the corresponding electric-field vectors $\mathbf{E}(t)$, vector potentials $\mathbf{A}(t) = -\int_0^t \mathbf{E}(t')dt'$, and the relevant electron trajectories and velocities between the ionization and recombination times, which correspond to the dominant quantum orbit SPA1, for the harmonic orders $n = 37$ (left panels) and $n = 109$ (right panels). Electron trajectories $\mathbf{r}(t)$ and velocities $\mathbf{v}(t)$ are calculated introducing our quantum-orbit solutions into the Newton equation of motion $\ddot{\mathbf{r}}(t) = -\mathbf{E}(t)$. For real times $t \in [\text{Re} t_{0s}, \text{Re} t_{1s}]$ they are given by

$$
\mathbf{r}(t) = \text{Re} \left[ \int_0^t \mathbf{A}(t')dt' - \int_{t_{0s}}^{t} \mathbf{A}(t')dt' + (t - t_{0s})\mathbf{k}_{st} \right], \quad \mathbf{v}(t) = \text{Re} \left[ \mathbf{k}_{st} + \mathbf{A}(t) \right],
$$

(5)
where the stationary electron momentum is \( k_{st} = -\int_{t_0}^{t_{rec}} dt' \mathbf{A}(t')/(t_1 - t_0) \). From the upper left subpanels of figure 2 we see that the electric field resembles that of a circular field or of a bichromatic circularly polarized field with co-rotating components (see, for example, [21]), for which HHG is negligible. However, in the time interval between the ionization and recombination times, denoted by I and R, respectively, there are approximately linear segments of the electric field vector, which can enable return of the electron to the parent core. The corresponding electron trajectories are shown in the upper right subpanels. The electron is ‘born’ at the time \( \text{Re} t_{0s} \) when the field is close to its maximum, few atomic units away from the nucleus, which is at the origin \((x, y) = (0, 0)\), and moves away from it, then turns around and moves back to the nucleus, where it recombines. The electron velocity, shown in the lower right subpanels, follows the shape of the vector potential, which is depicted in the lower left subpanels. For the \( n = 109 \) case vector potential at the recombination time is close to maximum. Important is that the electron velocity at the ionization time is small. The smaller is this velocity, the larger is the ionization probability. This explains why the corresponding high-harmonic intensity is large.

We also see that the behaviour of the electron velocity at the recombination time is different for the \( n = 37 \) and \( n = 109 \) cases: the corresponding curve approaches the point denoted by R from the left for the \( n = 37 \) case and from the top for the \( n = 109 \) case. This may explain large helicity asymmetry for the \( n = 37 \) case. Namely, it was found in [22] that the polarization of soft x-rays emitted in bichiral-laser-field-assisted electron-ion radiative recombination can be close to circular for low emitted x-ray photon energies and for a wide range of incident electron angles centered at \( j \cdot 360^\circ/(r + s) \) \((j \text{ integer})\). For example, for \( \omega - 3\omega \) bichiral field, for x-ray photon energies near 40\(\omega \) and incident electron angles near \( j \cdot 90^\circ \) the ellipticity is close to +1. For HHG process selection rules ensure that the ellipticity is exactly +1 or -1. Analyzing the electron velocities at the recombination time using quantum-orbit theory, we found that for \( n = 37 \) this angle \( \theta_{\text{rec}} \) is small \((j = 0 \text{ and } |\theta_{\text{rec}}| < 20^\circ)\). This favours the emission of low harmonics having...
ellipticity $\varepsilon_n = +1$ and large positive value of the helicity asymmetry parameter, exactly as observed in figure 1 (see also figure 3).

4. Focal-averaged numerical results

![Figure 3](image_url)

**Figure 3.** Focal-averaged results for the logarithm of the harmonic intensity (upper panels) and the helicity asymmetry parameter (lower panels) for HHG by $\omega-2\omega$ bicircular field with the fundamental wavelength 1300 nm. The results are presented in false colors as a function of the natural logarithm of the ratio of the component peak intensities, $\ln(I_2/I_1)$, and of the harmonic order $n$. Left panels: results for HHG by He atoms and the sum of the component peak intensities $I_1 + I_2 = 1.8 \times 10^{15}$ W/cm$^2$. Right panels: results for Ne atoms and the sum of the component peak intensities $I_1 + I_2 = 6 \times 10^{14}$ W/cm$^2$.

In figure 3 we present focal-averaged results, obtained by numerical integration, for HHG by $\omega-2\omega$ bicircular laser field with the fundamental wavelength 1300 nm. Results shown in the left panels are for He atoms and the sum of the component peak intensities $I_1 + I_2 = 1.8 \times 10^{15}$ W/cm$^2$. The ratio of the component intensities $I_2/I_1$ changes from $1/8$ to $8$. From the upper left panel we see that the harmonic intensity is much higher for $I_2 > I_1$. The cutoff position has maximum near $I_2 = 2I_1$ and slowly decreases with the increase of $I_2/I_1$. The helicity asymmetry parameter, shown in the lower left panel, is close to zero for harmonic order larger than $n = 50$. For $I_2 < I_1$ the helicity asymmetry parameter is close to $+1$ for $n < 50$. For $I_2 > I_1$ the interval of values of the helicity asymmetry parameter close to $+1$ is narrower and centered below $n = 40$. The reason why the helicity asymmetry parameter is close to $+1$ for low harmonic order is connected with the behaviour of the recombination matrix element. It was
shown in [22] that the polarization of soft x-rays emitted in bicircular-laser-field-assisted electron- 
ion radiative recombination can be close to circular for low emitted x-ray photon energies and 
for a wide range of incident electron angles centered at 0°, 120° and 240°. In addition, for low 
energies the imaginary parts of the saddle-point solutions are larger than in the high-energy 
case, which can also influence on the helicity asymmetry parameter.

In the right-hand panels of figure 3 we show focal-averaged results for HHG by Ne atoms 
and the sum of the component peak intensities \( I_1 + I_2 = 6 \times 10^{14} \, \text{W/cm}^2 \). The ratio \( I_2/I_1 \) 
changes from 1/7 to 7. Harmonic intensity presented in false colors as a function of the natural 
logarithm of the ratio of the component peak intensities and of the harmonic order \( n \) behaves 
similarly as for the example of HHG by He atoms, presented in left panels. This can be seen 
comparing the upper panels in figure 3. The cutoff value is larger in the He case due to larger 
peak intensity, but otherwise the results are qualitatively similar. However, the results for the 
helicity asymmetry parameter for Ne are very different: it is zero only in a small interval of 
harmonic orders, which is near \( n = 50 \) for \( I_2 < I_1 \) and which shifts to larger values of \( n \) with the 
increase of the ratio \( I_2/I_1 \). Above some critical value of \( n \) the helicity asymmetry parameter is 
close to \(-1\) and below this value it is close to \(+1\). The reason for positive values of the helicity 
asymmetry parameter for low harmonic order is related to the recombination matrix element, 
as in the case of He atoms. However, the negative value of the helicity asymmetry parameter for 
higher harmonic order is related to the \( p \) ground state of Ne atoms and the asymmetry in the 
\( m = +1 \) and \( m = -1 \) contributions to the quantum-mechanical time-dependent dipole \( \mathbf{d}_m(t) \). 
Detailed explanation and the corresponding semiclassical model are given in [15, 17, 18] and we 
will not repeat this here.

5. Conclusions

We have explored high-order harmonic generation by \( \omega-2\omega \) and \( \omega-3\omega \) bicircular laser fields 
for various ratios of their component intensities. We have found that even for much higher 
intensity of the second field component the high-harmonic intensity can be very high. This is 
illustrated by an example of HHG with He atoms, \( \omega-3\omega \) field and \( I_2 = 8I_1 \). The obtained results 
are explained using quantum-orbit theory and by identifying relevant electron trajectories and 
velocities. We have also explained why in this case low-order high harmonics having positive 
helicity are stronger. This is related to the recombination matrix element and the corresponding 
incident electron velocities at the recombination time.

For the \( \omega-2\omega \) bicircular field case we have analysed focal-averaged results for high-order 
harmonic intensity and helicity asymmetry parameter for a wide range of the ratios of the 
bicircular field component intensities. For He atoms, having the \( s \) ground state, helicity 
asymmetry parameter is close to zero except for low-order harmonics, where it is close to \(+1\). 
On the other hand, for Ne atoms, having the \( p \) ground state, the helicity asymmetry parameter 
can be large for higher harmonic orders and in a wider range of the driving field component 
intensities. It is negative for higher high-harmonic orders, while for low harmonics it is positive, 
similarly as in the case of He atoms. The helicity asymmetry for higher harmonic orders is 
related to the asymmetry in the \( m = +1 \) and \( m = -1 \) magnetic quantum number contributions 
to the ionization and recombination matrix elements, which appears for atoms having the \( p \) 
ground state (Ne, Ar, Kr and Xe; example for Ne atoms is presented).

High-order harmonic intensity for He and Ne has similar behaviour as a function of the 
harmonic order \( n \) and the ratio \( I_2/I_1 \). It is much higher for \( I_2 > I_1 \). The cutoff position has 
maximum near \( I_2 = 2I_1 \) and slowly decreases with the increase of \( I_2/I_1 \). It also increases with 
the increase of the sum of the bicircular field component intensities. The presented results can 
serve physicists to design their experiments for exploration of the chirality sensitive processes.
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