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Generation of elliptically polarized soft x rays using high-order harmonic generation with orthogonal two-color laser fields

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Abstract. High-order harmonic generation by orthogonally polarized two-color (OTC) laser fields is analysed using strong-field approximation and quantum-orbit theory. Results for the field components frequency ratio of 2:1 and 3:1 are presented and compared. We have shown that, depending on the relative phase between the field components, the shape of the high-harmonic spectrum can be very different from that obtained by a monochromatic linearly polarized laser field. It is also shown that it is possible to generate elliptically polarized high-order harmonics with very high photon energies using OTC laser field with the frequency ratio of 3:1 and a long fundamental wavelength. An effective relative phase control of the harmonic emission is demonstrated. The obtained results are explained using the quantum-orbit theory.

1. Introduction

For various applications, such as the investigation of magnetic materials or chiral molecules, elliptically polarized coherent soft x rays are a mandatory tool [1]. It is very desirable to have tabletop schemes for their production [2]. In the high-order harmonic generation (HHG) process, energy is absorbed from the laser field and re-emitted in the form of a high-energy photon, which might have the desired properties. However, a linearly polarized laser field generates only linearly polarized harmonics. Hence, in order to generate elliptically polarized soft x rays, we need more a complex laser field configuration. One such configuration, proposed in 1995 [3], is the so-called bicircular laser field, which consists of two coplanar counter-rotating circularly polarized fields of different frequencies equal to integer multiples of a fundamental frequency ω . The high-order harmonics generated by a bicircular laser field are circularly polarized with helicities that alternate from one harmonic order to the next. Combining a group of such harmonics one obtains a pulse in the form of a star-like structure consisting of three linearly polarized pulses, which are rotated versus one another by 120 degrees [4] (for a frequency ratio of 2:1). For HHG by atoms having a p ground state the harmonics of one or the other helicity are stronger and it is possible to generate an elliptical or even circular attosecond pulse train [5, 6, 7]. Theoretical



investigations of HHG by a bicircular field were presented in [8] for a zero-range-potential atom and in [9] based on the strong-field approximation (SFA) and quantum-orbit theory (see also [10] and references therein).

In this paper we will consider another bichromatic field configuration – the orthogonally polarized two-color (OTC) laser field. This field consists of two orthogonal linearly polarized fields with a frequency ratio of 2:1 or 3:1 and fixed relative phase. The frequency ratio 2:1, i.e. the ω – 2ω OTC field, has been used for many applications (see [11] and references therein), while the ω – 3ω OTC field has received almost no attention at all (we have found only an old experimental paper [12]; first theoretical results for this field can be found in [8]). For HHG by the ω – 2ω OTC field the harmonics are linearly polarized: odd harmonics are polarized in the direction of the ω field component, while the even harmonics are linearly polarized in the direction of the 2ω component. On the other hand, for the ω – 3ω OTC field only odd harmonics are emitted, which have elliptical polarization. For the investigation of HHG by the ω – 2ω OTC field, the discovery of the phase-dependent enhancement of the HHG yield was crucial [13]. (see also [14, 15]; for further developments, see references in [11]).

In the present work we will analyze HHG by neon atoms and ω – 2ω or ω – 3ω OTC fields. In section 2 we introduce the strong-field approximation and quantum-orbit theory, applied to the OTC fields. In section 3 we present our numerical results, while our conclusions are given in section 4. We use the atomic system of units.

2. Theory

The electric-field vector of our OTC field is

$$\mathbf{E}(t) = E_1 \cos(\omega t) \hat{\mathbf{e}}_x + E_2 \sin(j\omega t + \phi) \hat{\mathbf{e}}_y, \quad j = 2, 3. \quad (1)$$

Here the unit vectors $\hat{\mathbf{e}}_x$ and $\hat{\mathbf{e}}_y$ span the xy plane and we consider ω – 2ω and ω – 3ω OTC fields with the relative phase ϕ between the two field components.

We calculate our results using the SFA applied to the HHG process [16, 17, 18, 19, 20, 10]. The n th harmonic intensity

$$I_n = \frac{(n\omega)^4}{2\pi c^3} (|T_n^x|^2 + |T_n^y|^2) \quad (2)$$

is related to the components T_n^x and T_n^y of the T -matrix element, which are calculated as the following integral over the recombination time t :

$$\mathbf{T}_n = T_n^x \hat{\mathbf{e}}_x + T_n^y \hat{\mathbf{e}}_y = \int_0^T dt \mathbf{d}(t) e^{in\omega t} / T, \quad T = 2\pi/\omega. \quad (3)$$

The time-dependent dipole $\mathbf{d}(t)$ implies an additional integral over the ionization time t_0 :

$$\mathbf{d}(t) = -i \left(\frac{2\pi}{i} \right)^{3/2} \int_{-\infty}^t \frac{dt_0}{(t - t_0)^{3/2}} \langle \psi_g | \mathbf{r} | \mathbf{k}_{\text{st}} + \mathbf{A}(t) \rangle \langle \mathbf{k}_{\text{st}} + \mathbf{A}(t_0) | \mathbf{r} \cdot \mathbf{E}(t_0) | \psi_g \rangle e^{iS_{\text{st}}}. \quad (4)$$

Here $\mathbf{A}(t) = -\int^t dt' \mathbf{E}(t')$, $\mathbf{k}_{\text{st}} = -\int_{t_0}^t dt' \mathbf{A}(t') / (t - t_0)$ is the stationary momentum, and ψ_g is the p ground-state atomic wave function of the neon atom modelled by a linear combination of Slater-type orbitals. The stationary action is $S_{\text{st}} \equiv -I_p(t - t_0) - \int_{t_0}^t dt' [\mathbf{k}_{\text{st}} + \mathbf{A}(t')]^2 / 2$.

Rather than by numerical integration, the above integrals can be evaluated using the saddle-point method, which leads to the conditions $\partial S_{\text{st}} / \partial t_0 = 0$ and $\partial(S_{\text{st}} + n\omega) / \partial t = 0$. From these conditions we obtain the following system of equations

$$\frac{1}{2} [\mathbf{k}_{\text{st}} + \mathbf{A}(t_0)]^2 = -I_p, \quad n\omega = \frac{1}{2} [\mathbf{k}_{\text{st}} + \mathbf{A}(t)]^2 + I_p, \quad (5)$$

for the complex times t_0 and t . The energy of the emitted n th harmonic is $n\omega$ and I_p is the ionization potential of the neon atom. Physically, the conditions (5) express energy conservation at the ionization and recombination times. For a linearly polarized laser field, we classified the saddle-point solutions by the multi-index $(\alpha, \beta, m) \equiv s$ [21]. The index $m = 0, 1, 2, \dots$ determines the approximate length of the travel time $\text{Re}(t_s - t_{0s})$ in multiples of the laser period T . The index $\beta = \pm 1$ distinguishes the two pairs of solutions within one optical cycle, while the longer and shorter orbits of each pair are discriminated by the index $\alpha = \pm 1$. The same classification can be used for the saddle-point solutions for the OTC fields [11].

The T -matrix element in the saddle-point approximation can be written as $\mathbf{T}_n = \sum_s \mathbf{d}(t_s) e^{in\omega t_s}$. From (5) we see that, for the $\omega-3\omega$ OTC field (1), if $\{t_{0s}, t_s\}$ is a solution of the saddle-point equations then so is $\{t_{0s} + T/2, t_s + T/2\}$. For this field we have $\mathbf{d}(t_s + T/2) = -\mathbf{d}(t_s)$ and $\mathbf{T}_n = \sum_s \mathbf{d}(t_s) e^{in\omega t_s} (1 - e^{in\pi})$, so that only odd harmonics are emitted and they have both $\hat{\mathbf{e}}_x$ and $\hat{\mathbf{e}}_y$ components. Therefore, the emitted high harmonics are elliptically polarized. The ellipticity of the n th harmonic is

$$\varepsilon_n = \text{sgn}(\xi_n) \sqrt{\frac{1 - \sqrt{1 - \xi_n^2}}{1 + \sqrt{1 - \xi_n^2}}}, \quad \xi_n = \frac{\text{Im}(2T_n^{x*} T_n^y)}{|\mathbf{T}_n|^2}. \quad (6)$$

The electron trajectories (quantum orbits) $\mathbf{r}(t)$ and velocities $\mathbf{v}(t)$ [22, 23, 24, 25, 26], are calculated by introducing the saddle-point solutions t_{0s} and t_s into the following solutions of the Newton equation of motion $\ddot{\mathbf{r}}(t) = -\mathbf{E}(t)$:

$$\mathbf{r}(t_R) = \text{Re} \left[\int_{t_{0s}}^{t_R} \mathbf{A}(t') dt' + (t_R - t_{0s}) \mathbf{k}_{\text{st}} \right], \quad \mathbf{v}(t_R) = \text{Re} [\mathbf{k}_{\text{st}} + \mathbf{A}(t_R)]. \quad (7)$$

Here the time $t_R \in [\text{Re } t_{0s}, \text{Re } t_s]$ is real and we will project $\mathbf{r}(t_R)$ and $\mathbf{v}(t_R)$ into the real plane.

3. Results

We will present numerical results for HHG by Ne atoms and an OTC field having the fundamental wavelength 1300 nm and the first component intensity $I_1 = E_1^2 = 2 \times 10^{14} \text{ W/cm}^2$. We will compare the results for the $\omega-2\omega$ and $\omega-3\omega$ OTC fields. In order to see if the saddle-point classification by the multi-index (α, β, m) , introduced for a monochromatic linearly polarized field in [21], is applicable to the OTC fields, we compare the SFA and saddle-point results for various intensities $I_2 = E_2^2$ of the second field component, from a few percent of I_1 to $I_2 = I_1$. We will also present saddle-point solutions for complex ionization and recombination times as well as examples of quantum orbits.

In figure 1 we present SFA results for equal component intensities. From the upper left panel we see that, for the $\omega-2\omega$ OTC field and the phase $\phi = 0$ rad, the spectrum does not exhibit the typical behaviour known from the monochromatic field, i.e. a flat plateau with an abrupt cutoff. Instead, having a typical fast decrease for the first few harmonics followed by a flat part for the low harmonics, after the harmonic $n = 40$ the spectrum exhibits an intensity increase, forming a semicircular shape with its maximum intensity near $n = 100$, and then decreases up to the harmonic $n = 150$ by which order the harmonic intensity is negligible. In the region near the maximum ($n = 100$) the odd harmonics are substantially stronger than the even harmonics. Odd and even harmonics are linearly polarized but in mutually perpendicular directions. In the same panel we also present the results obtained for the relative phase $\phi = 1$ rad. We see that in the high-energy region now the even harmonics are stronger. The spectrum is structured and exhibits a sharp peak near the cutoff at the harmonic order $n = 140$.

In the upper right panel, we present harmonic spectra for the $\omega-3\omega$ OTC field for several different values of the relative phase. Only odd harmonics are emitted and they are elliptically

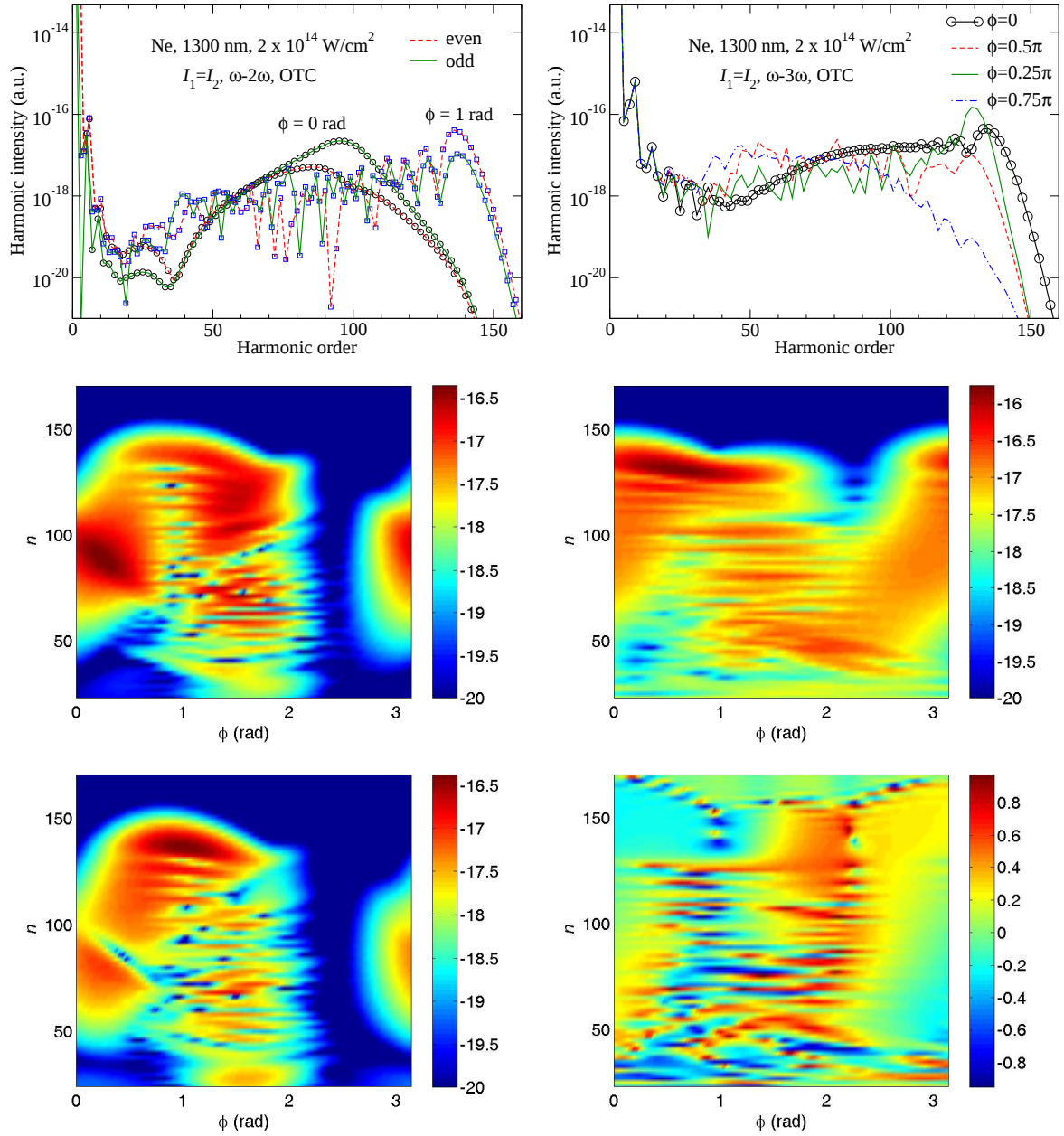


Figure 1. SFA results for HHG by Ne atoms and $\omega-2\omega$ (left panel) and $\omega-3\omega$ (right panel) OTC fields having equal component intensities $I_1 = I_2 = 2 \times 10^{14} \text{ W/cm}^2$ and wavelength 1300 nm. Top panels: Harmonic intensities as functions of the harmonic order for the relative phases $\phi = 0 \text{ rad}$ and $\phi = 1 \text{ rad}$ (left) and $\phi = k\pi/4 \text{ rad}$, $k = 0, 1, 2, 3$ (right panel). Middle and bottom panels: the results are presented in false colors as a function of the relative phase ϕ and harmonic order n . Middle (bottom) left panel: Logarithm of the harmonic intensity of odd (even) harmonics. Middle (bottom) right panel: Logarithm of the harmonic intensity (harmonic ellipticity).

polarized. For the relative phase $\phi = 0 \text{ rad}$ the spectrum forms a characteristic flat plateau with a sharp cutoff. The form of the plateau and the cutoff depend on the relative phase (for

$\phi = 0.75\pi$ rad there are no plateau and cutoff at all). For a monochromatic linearly polarized field, the harmonic intensity in the plateau exhibits sharp oscillations. This is explained by the interference of two (or more) dominant quantum orbits. However, for the ω - 3ω OTC field with $\phi = 0$ rad the plateau is flat (except in the cutoff region) which indicates that only one quantum orbit is dominant (the same holds for the ω - 2ω spectrum for $\phi = 0$ rad in the left panel).

Before we investigate this, in the remaining panels of figure 1 we explore in more detail how our results depend on the relative phase. In the left middle and bottom panels, for the ω - 2ω OTC field, we present the harmonic intensity (in atomic units, on a logarithmic scale and in false colors) as a function of the relative phase ϕ and harmonic order n . We see that both for odd (middle left panel) and even (bottom left panel) harmonics there are regions in which the harmonic intensity exhibits a sharp maximum. For the odd harmonics the corresponding harmonic order is slightly below $n = 100$, while the corresponding harmonic phase is slightly larger than zero. On the other hand, for the even harmonics the sharp maximum is near the harmonic order $n = 140$ and the phase near $\phi = 1$ rad (this behavior is better visible on a linear false-color scale). Therefore, a very efficient phase control of the HHG process by OTC fields is possible. This is even more visible for the ω - 3ω OTC field for which the results are shown in the right middle panel: the harmonic intensity exhibits a sharp peak (much sharper and having much higher intensity than in the ω - 2ω case) near $n = 140$ for a wide interval of the relative phase between 0 rad and 1.2 rad. For practical applications it is important that the harmonics generated by the ω - 3ω OTC field are elliptically polarized. The ellipticities are plotted in the right bottom panel of figure 1. We see that there are regions where both the harmonic ellipticity and intensity are large.

In figure 2 we compare the spectra of high harmonics generated by OTC fields calculated using the SFA (left panels) and the saddle-point method (right panels). The intensity of the first component is $I_1 = 2 \times 10^{14}$ W/cm², while the second-component intensity changes from zero to 30% of I_1 , as denoted in the upper left corner of each panel. For the ω - 2ω OTC field and low intensity of the second component the spectrum is similar to that generated by a monochromatic linearly polarized field, characterized by an oscillatory plateau and an abrupt cutoff. From the upper right panel we see that there are four pairs of partial saddle-point contributions to the harmonic yield. They are presented by solid and dashed lines having different colors for different second-component intensities (the contributions of the dashed lines should be disregarded after their cutoffs). When the second-component intensity increases the contributions of the longer quantum orbits, which contribute to lower harmonic orders, become suppressed. This is particularly pronounced for the case $I_2 = 30\%I_1$ (blue lines). In this case, only one quantum orbit contributes (blue solid line) and the partial contribution of this orbit to the harmonic yield has exactly the semicircular shape which we noticed in figure 1. Comparing the blue curves in the left and right upper panels in figure 2 we conclude that the saddle-point result obtained using the contribution of only one quantum orbit is in excellent agreement with the results of the numerical SFA integration. It should be noticed that the SFA results for the even and odd harmonics are different, which is responsible for the oscillations in the SFA curves, while in the saddle-point method the harmonic order is a continuous parameter. Similar results for the ω - 3ω OTC field are presented in the lower panels of figure 2. We see that, when the second-component intensity is increasing up to $0.3I_1$, the contribution of only one quantum orbit becomes dominant. This partial contribution (blue line) completely determines the shape of the HHG spectrum and agrees excellently with the SFA result presented in the lower left panel. The dominance of only one quantum orbit explains why the harmonic plateau is smooth and practically flat.

In [21] for a monochromatic linearly polarized field we classified the saddle-point solutions by the multi-index (α, β, m) . In figure 2 we exhibited the partial contributions to the harmonic yield of eight such solutions (or four pairs of solutions (β, m) having $\alpha = \pm 1$; the solutions

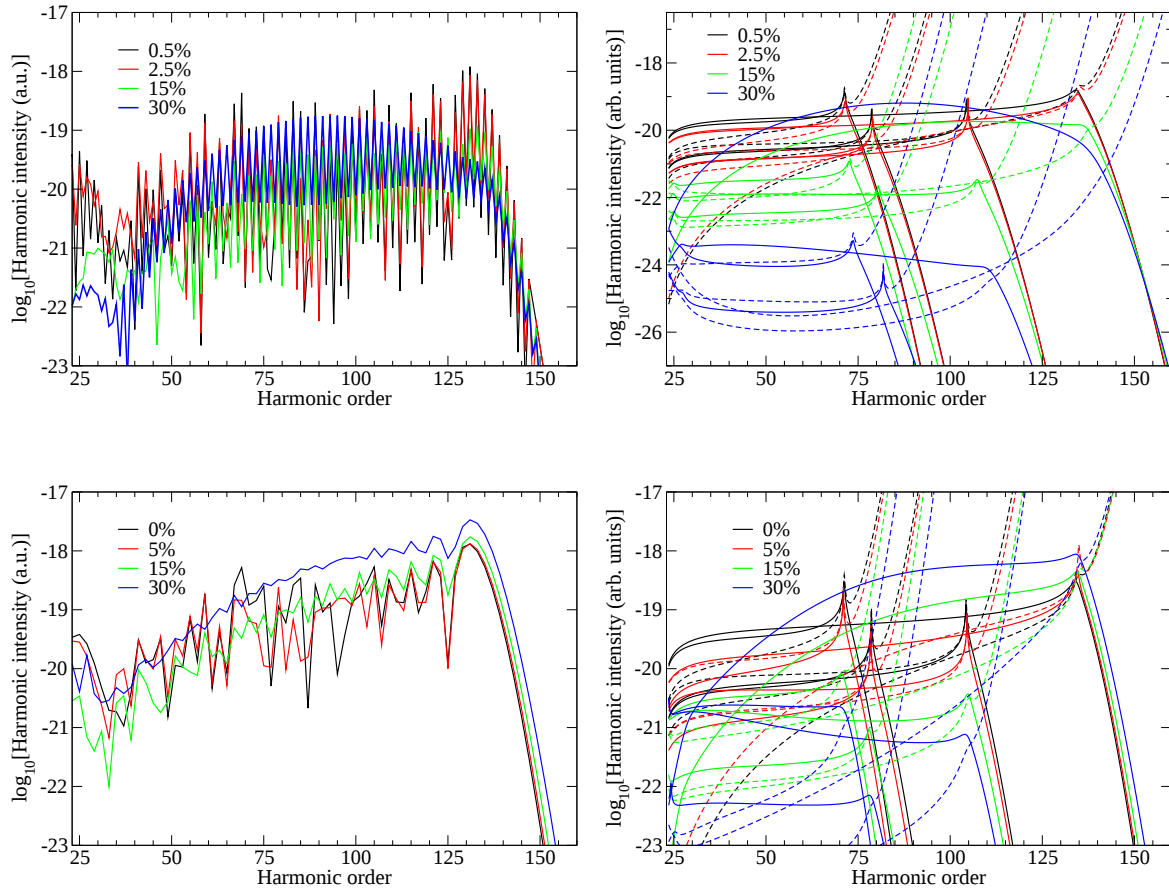


Figure 2. Comparison of harmonic spectra calculated by the SFA (left panels) and by the saddle-point-method (right panels) for Ne atoms and $\omega-2\omega$ (upper) and $\omega-3\omega$ (lower panels) OTC fields having wavelength 1300 nm, the first component intensity $I_1 = 2 \times 10^{14} \text{ W/cm}^2$ and the relative phase $\phi = 0 \text{ rad}$. The intensity of the second component is given in percent of the intensity I_1 , as denoted in the legends.

$\alpha = 1$ and $\alpha = -1$ are distinguished by the solid and dashed lines). In figure 3 we present the corresponding ionization times t_0 and recombination times t in the complex plane. We can see how these solutions move in the complex plane when the second OTC-field component-intensity increases. Our way of presentation is the same as in [21]: the harmonic order changes along each curve from a small to a large value beyond the cutoff. At the cutoff the two curves from the pair $\alpha = \pm 1$ approach each other and the cutoff harmonic order corresponds to the point of the closest approach. Thereafter, the curves move in opposite directions and the contribution to the harmonic yield of one of them (denoted by the dashed line) should be dropped since it is divergent after the cutoff (this problem is cured in the uniform approximation). We noticed that, upon an increase of the second-component intensity, the solutions travel and may rotate in the complex plane so that the “dashed” and the “solid” solutions interchange. This usually happens for $I_2 > 0.05I_1$.

In order to further explain the numerical results for the HHG spectra, we apply quantum-orbit theory. In figure 4 we separately present the results for the $\omega-2\omega$ (left panel) and $\omega-3\omega$ (right panel) OTC fields. In the upper (lower) left subpanel we show the corresponding electric-field

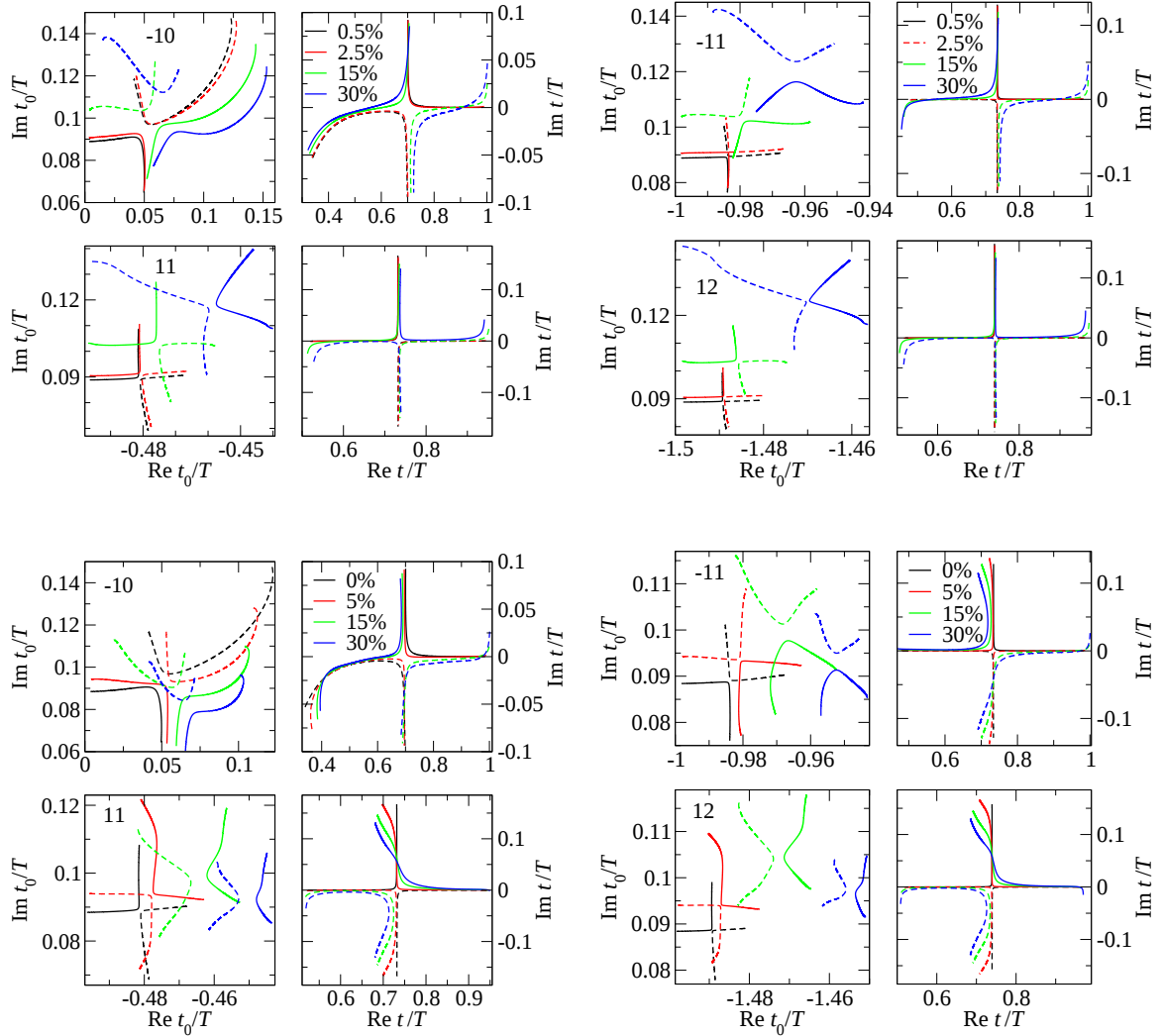


Figure 3. Saddle-point solutions for the complex times t_0 and t in units of the optical period $T = 2\pi/\omega$ for HHG of Ne atoms by $\omega-2\omega$ (upper) and $\omega-3\omega$ (lower panels) OTC fields having the wavelength 1300 nm, the first-component intensity $I_1 = 2 \times 10^{14} \text{ W/cm}^2$ and the relative phase $\phi = 0$ rad. The second-component intensity is given in percentages of the intensity I_1 and the corresponding solutions are presented by different colors, as denoted in the legends. The solutions $(\alpha, \beta, m) = (\pm 1, -1, 0), (\pm 1, 1, 1), (\pm 1, -1, 1), (\pm 1, 1, 2)$ are presented, with the index (β, m) indicated, for each pair of subpanels, in the upper left corner of the subpanel that corresponds to the ionization time t_0 (the right-hand subpanel corresponds to the recombination time t). The contributions of the solutions represented by dashed lines should be dropped after the cutoff.

vector $\mathbf{E}(t)$ (vector potential $\mathbf{A}(t)$), while in the upper (lower) right subpanel we present the electron trajectory (velocity) from the ionization time (marked on the trajectory by the letter I) to the recombination time (letter R). We display only the dominant shortest trajectory. The electron is “born” at the “exit of the tunnel” a few atomic units away from the nucleus and returns almost exactly to the nucleus. The shape of the trajectory is close to linear, the electron follows a line in the negative x direction, turns around and returns to the nucleus following a

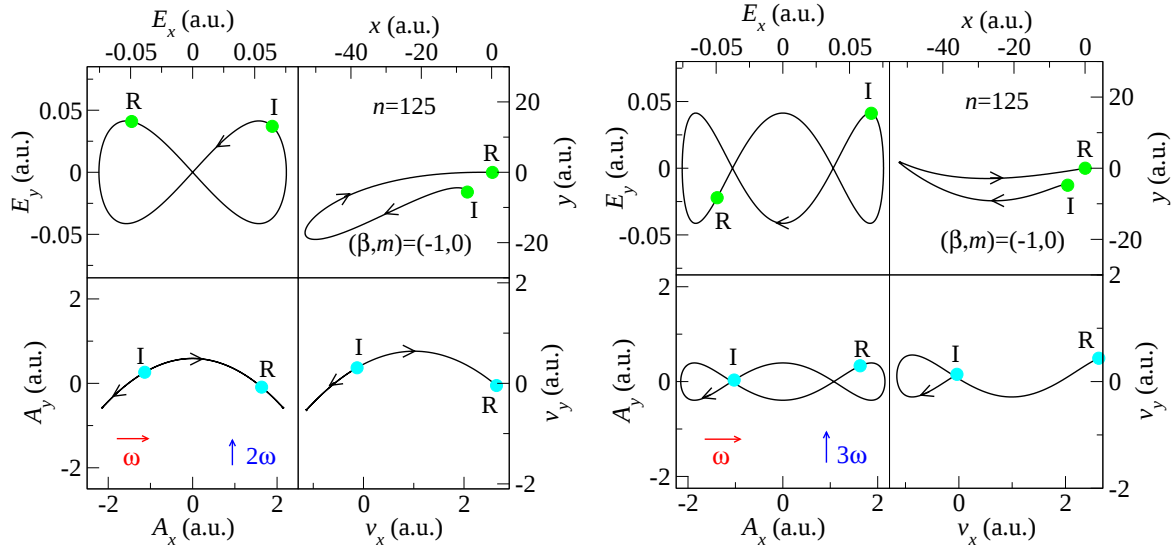


Figure 4. The $\omega-2\omega$ (left panel) and $\omega-3\omega$ (right panel) OTC electric-field vector $\mathbf{E}(t)$ (upper-left subpanels), vector potential $\mathbf{A}(t)$ (lower left), electron trajectory $\mathbf{r}(t)$ (upper right), and velocity $\mathbf{v}(t)$ (lower right) between the ionization time (I) and recombination time (R). The results are for Ne atoms, the fundamental laser wavelength 1300 nm, the component intensities $I_1 = 2 \times 10^{14}$ W/cm² and $I_2 = 0.3I_1$ and the relative phase $\phi = 0$ rad. Harmonic order is $n = 125$ and the results for the solution $(\beta, m) = (-1, 0)$ are presented.

line, which is almost parallel to the first part of the electron trajectory but has the opposite direction. This is similar to one-dimensional trajectories, which are characteristic for ionization by a linearly polarized field, or to the shortest trajectories for the bicircular-field case, which also approximately follow a straight line ([10] and references therein). For the $\omega-3\omega$ OTC field the electron velocity at the ionization time is almost zero so that the corresponding ionization probability and the harmonic intensity are high. This is in agreement with the numerical SFA results (compare the blue curve in the lower left panel in figure 2). On the other hand, for the $\omega-2\omega$ OTC field the corresponding velocity at the ionization time is relatively large. Having in mind that the ionization probability decreases exponentially with the square of this initial electron velocity, we expect that the corresponding harmonic intensity is also low. The upper left panel in figure 2 confirms that this is true. The results presented in the other subpanels of figure 4 can also help to explain the numerical results obtained. For example, the electric field is strong at the ionization time, the vector potential and the electron velocity at the recombination time are large, etc. Therefore, the quantum-orbit formalism is very helpful in explaining in detail the HHG process in OTC fields.

4. Conclusions

High-order harmonic generation by orthogonally polarized two-color laser fields is investigated theoretically using the strong-field approximation and quantum-orbit theory. Results for $\omega-2\omega$ and $\omega-3\omega$ OTC fields are compared. In both cases the harmonic spectra can be controlled by changing the relative phase between the field components. For comparable intensities of the two field components and appropriate phases, the spectrum for the $\omega-2\omega$ OTC field has a semicircular shape very different from the usual shape for a monochromatic linearly polarized laser field, which is characterized by an oscillatory plateau followed by a sharp cutoff. However, for the $\omega-3\omega$ OTC field, for a particular interval of the relative phases, it is possible to achieve

the usual plateau-cutoff shape of the spectrum. Furthermore, the plateau can be flat. This behaviour of the HHG spectrum can be explained using quantum-orbit theory, which shows that in this case only one quantum orbit becomes dominant. The flat plateau for the $\omega-3\omega$ OTC field can be important for applications. Namely, combining a group of harmonics in the plateau one can obtain an attosecond pulse train with better characteristics than in the case of a monochromatic linearly polarized field.

High harmonics generated by the $\omega-3\omega$ OTC field are elliptically polarized. We presented results for HHG by Ne atoms and a wavelength of 1300 nm. If we want to increase the energy of the high-harmonic photon into the soft x-ray region we should increase the laser wavelength and intensity. The highest energy can be achieved using He atoms, which have an s ground state. We have recently shown [11] that it is possible to generate elliptically polarized high-order harmonics with photon energies in the keV region using He atoms and an $\omega-3\omega$ two-color laser field with mutually orthogonal linearly polarized components and a fundamental wavelength of 2200 nm.

Acknowledgments

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