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# Generation of even and odd harmonics in the XUV region with controlling the relative delay and polarization of two-color fields

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## ABSTRACT

We report high harmonic generation in the extreme ultraviolet (XUV) region by using two-color laser fields at the wavelengths of 800 nm and 400 nm. With a Ti: sapphire femtosecond laser at a 10 Hz repetition rate efficient high harmonic generation was obtained in an argon gas medium with the IR field ( $\omega$ ) and its second harmonic (2 $\omega$ ) with parallel and perpendicular polarizations. The relative delay between the pulses is controlled via a Michelson interferometer arrangement. In addition to the odd high harmonic spectrum with frequencies  $(2n+1)\omega$ , where n - integer number, also even spectral components 2no are obtained, when the fields of the two colors are temporally overlapped and interact with the argon gas. Due to this overlap, the yield of odd harmonics can be increased as well. The calculations of electron trajectories within the semiclassical approach show differences in the action of the fields with parallel and perpendicular polarizations. For the former, there exist broad time intervals for returning electrons, and in the latter case there are only narrow intervals for possible solutions for each of the electron returns to the parent ion for a fixed phase difference between the  $\omega$  and  $2\omega$  fields. By varying the relative delay between the two laser fields it is possible to spectrally enrich and enhance the produced XUV radiation or suppress it. The conclusions of the semi-classical approach were confirmed by calculations of the harmonic intensities within the strong field dipole approximation.

### 1. Introduction

Interaction of a strong laser field with atoms leads to the generation of harmonics of the fundamental field due to the process called high harmonic generation (HHG), and the spectrum of the produced radiation can extend to vacuum ultraviolet, extreme ultraviolet (XUV) and even soft x-rays spectral regions. This novel light source delivers pulses with a duration from femtosecond to attoseconds [1]. The HHG finds many potential applications in science and technology, from attosecond physics [2] to microscopic imaging [3–6] and from interferometry [7] to the exploration of atomic and molecular structures [8–10]. Imaging of electronic density distributions [11] with the generated attosecond pulses [12,13] was demonstrated by different research groups, proving the unique properties of the HHG sources. However, the HHG process suffers from its low conversion efficiency of the fundamental field into the XUV radiation. There have been several methods developed to diminish this drawback for applications of HHG as a practically efficient and useful source of the XUV radiation. Approaches using two gas cells or mixing two different gases were experimentally tested [14–17]. Also

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quasi-phase-matching between the fundamental laser field and the generated harmonics was employed to improve the harmonic yields [18]. Another approach to significantly improve the efficiency of the HHG process is to use a two-color laser field, which can be a combination of the fundamental laser field (IR,  $\omega$ ) and its second harmonic (SH,  $2\omega$ ) [19–21]. Such two-color field not only can improve the conversion efficiency into high harmonics, but it also results in the generation of even harmonics besides the odd ones, when the inversion symmetry of the driving field is broken [22]. An enhancement and spectral extension of generated harmonics can be achieved with spatially inhomogeneous fields [23]. A possibility to selectively manipulate the spectrum of high harmonics with two- and three-color fields was also demonstrated [24].

The three-step model [25–27] well explains HHG phenomena. In the first step, an electron is tunnel-ionized, when the Coulomb potential confining the electron is tilted by the strong laser field [27]. In the second step, the electron gains kinetic energy, while propagating in the oscillating laser field. In the third step, the electron releases its energy in the form of an energetic photon, recombining with its parent atom. The energy of the released photon is determined by the sum of the gained kinetic energy and the ionization potential of the gas medium used. This three-step model formulated for the linearly polarized one-color field explains the emission of odd harmonics. We note that in the case of an elliptical polarization of the field, the electron can miss its parent atom, and consequently, harmonic generation, in this case, will be strongly reduced. However, with specially tailored circularly polarized two-color fields HHG also has been observed [28,29]. The frequencies of the harmonic spectra were extended to the XUV and even to the soft X-ray region when using driving fields with longer wavelengths [19,29]. Some spectral components in the XUV region, inaccessible with a driving field at one carrier frequency, become accessible by mixing different carrier frequencies in the driving field [30,31]. Moreover, it was demonstrated that by changing the delay of a weak 1300 nm mid-infrared (MIR) pulse relative to an intense multicycle 800 nm laser pulse it was possible to cause a spectral shift of harmonic peaks [32].

Two-color laser fields have been used in several studies for generation of odd and even harmonic orders, enhancement and control of their divergence. With orthogonally polarized two-color field the control of the divergence of HHG and an improvement of the harmonic yield were demonstrated [33]. A weak second harmonic field enabled the reconstruction of electron transition times [34–36]. Moreover, by applying the second harmonic field the selection between the long or short electron trajectories is possible [37, 38]. Two-color HHG effectively improves plateau harmonics yields [39]. For different configurations of multi-color fields the increase of the HHG yield was also observed [24,40–49].



**Fig. 1.** (a) HHG experimental setup with a variable relative time delay of the driving fields. TS: translation stage, M: mirror, BS: beam splitter, FL: focusing lens, BBO: beta barium borate doubling crystal, HWP: half-wave plate, GJ: gas jet, MCP: microchannel plate. (b) Schematic of the relative delay between the two produced pairs of double pulses centered at 800 nm ( $\omega$ , red color) and at 400 nm field ( $2\omega$ , blue color) that were used for the HHG. Two series of measurements were taken, one with and another without the HWP (details in the text). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article).

In this study, we present additional results on the HHG process with two-color laser fields and investigate the generation of high harmonics when varying the relative delay between the two-color laser fields with parallel and orthogonally polarized fields. In addition to odd harmonics produced by an 800 nm driving laser field, even-order harmonics are generated when the fundamental field and its second harmonic (400 nm) are focused into an argon gas medium and spatiotemporally overlapped in the interaction region. This becomes possible by breaking the symmetry of the superposition field. HHG with two-color fields brings additional parameters, such as the relative orientation of polarizations, used wavelengths and intensities, and the relative delay of the two fields, allowing for additional control of the XUV output. By analyzing electron trajectories, we show that the two-color HHG with crossed polarizations realizes synchronization of the return and recombination of the ionized electrons resulting in a higher yield of XUV radiation and creating conditions for generation of ultrashort (attosecond) pulses.

#### 2. Experimental setup

The experimental setup is presented in Fig. 1 (a). The experiments were performed with a chirped-pulse amplified Ti: Sapphire laser system by using pulses with a duration 50 fs, central wavelength 800 nm, and output energy per pulse up to 6 mJ at a 10 Hz repetition rate. The laser output is split into two arms by using a beam splitter. One arm (we call it "fixed") has a fixed optical path, while for the other "variable" arm the delay can be varied with a motorized translation stage (TS), changing the delay  $\Delta t$  (see Fig. 1(b)). The spatiotemporal overlap of the pulses from the two arms and their zero delays after the BS-2 were carefully established by looking at the changing interference pattern, as the delay produced by the translation stage was varied.

These overlapped beams are focused with a ~40 cm focal length lens into a gas jet produced with a ~0.5 mm thick nickel tube; more details on this arrangement can be found in Refs. [50–52]. The 400 nm pulses with polarization perpendicular to the 800 nm radiation are directly produced with a BBO crystal placed after the focusing lens, while to produce the parallel polarization of the 400 nm and 800 nm fields the HWP is appropriately rotated. Consequently, there were two pairs of pulses entering the chamber and interacting with the gas jet, each including IR and SH fields generated after the BBO crystal. A fixed delay between the IR and SH pulses of  $\Delta t_0 \sim 0.5$  ps for perpendicular polarizations and  $\Delta t_0 \sim 0.8$  ps for parallel polarization is introduced due to the differences of the refractive indexes of the red and blue pulses in the entrance window and the HWP (Fig. 1 (b)). In the focal region, the intensity of the  $\omega$ beam is  $\sim 3 \times 10^{14}$ W/cm<sup>2</sup>, and the intensity of the 2 $\omega$  beam is  $\sim 7 \times 10^{13}$ W/cm<sup>2</sup>.

The pressure in the gas jet was set to  $\sim$ 50 mbar, and the pressure at the micro-channel plate (MCP) detector was reduced to  $\sim 10^{-6}$ mbar, thus minimizing the reabsorption of the produced XUV radiation. The latter was spectrally dispersed by the grating of an XUV spectrometer (McPherson, 248/310 G). Then an angular segment of the dispersed radiation was detected by an imaging MCP. The detected harmonics were imaged on a fluorescent screen, and the images were re-captured by a CCD camera and digitized. All the devices were controlled by a LabVIEW program.

## 3. Experimental results

The experiment is performed to investigate the role of the relative time delay between the fields of two colors on the HHG process. Fig. 2 shows in false colors the spectral density plot of the yields of high harmonics for different relative delays in the range of the delay times between the two pairs of pulses from -0.2 ps to +1 ps. The fields for  $\omega$  (IR) and 2 $\omega$  (SH) pulses are orthogonally polarized. The spectra of odd high harmonics from 13H to 21H appear as vertical stripes. The strong variations of the spectral density with the changing delay, especially close to zero delay (the horizontal stripes) are due to constructive and destructive interference of the two pulse pairs. To overlap the  $\omega$  and 2 $\omega$  pulses from the different pairs of pulses a delay between the two optical arms should be introduced



Fig. 2. Spectral density plot for different relative delays in the range of the delay times from -0.2 ps to +1 ps. HHs of odd orders appear as vertical stripes. HHs of even orders appear at delay ~0.5 ps, corresponding to the temporal overlap of the red and blue pulses;  $\omega$  and  $2\omega$  fields were orthogonally polarized, and a time step of 2.33 fs was used for the delay. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article).

#### M. Sayrac et al.

equal to the delay between  $\omega$  and  $2\omega$  pulses in a single pair. The plot in Fig. 2 shows that the generation of even high harmonics (HHs) takes place at the delay  $\Delta t = \Delta t_0 \sim 0.5$  ps corresponding to the overlap of the  $2\omega$  pulse from the first pair with the  $\omega$  pulse from the second pair (Fig. 1(b)).

In another measurement, we controlled the polarization of the fundamental field by placing the HWP after the BBO crystal. The HWP was rotated to change the polarization of the fundamental field, so that both fields have parallel polarizations. Fig. 3 shows the spectral density of the XUV output for parallel polarized fields at different delays. When both the IR and the SH fields were temporally overlapped at  $\sim 0.8$  ps time delay, the generation of even HHs of orders from 14 to 22 was observed. The larger time delay for the  $\omega$  and  $2\omega$  pulses to overlap is required in this case, because the HWP introduces an additional delay between the pulses of two colors. The comparison of the spectral densities for orthogonal and parallel polarizations (Fig. 4) shows that the yield of the XUV radiation is higher for the orthogonal case.

To see the effect of the relative phase of the IR and SH fields, the high harmonics spectra were measured at a discrete set of delays with the small delay increments, corresponding to a relative phase change  $\phi$  of the 400 nm light in respect to the 800 nm light (see Eq. (1)) by  $\Delta \phi = \pi/4$ , as shown in Fig. 5(a). This data exhibit variations of the HHs yields showing maxima each half period with the relative magnitude of these variations increasing for higher-order harmonics (Fig. 5(b)). The origin of these variations is discussed in the next section considering classical electron dynamics in a two-color field.

### 4. Simulations of the electron behavior in a two-color field

To get some insights into the processes behind the observed spectra, we considered the motion of electrons in the two-color field after ionization. We calculated classically the motion of an electron within 1–2 periods of a relatively long pulse containing several tens of periods (as in our case), so the field can be approximated as

$$\overline{E(t)} = \overline{E_{\omega}} \sin(\omega t) + \overline{E_{2\omega}} \sin(2\omega t + \phi), \tag{1}$$

where  $\overrightarrow{E_{\omega}}$  and  $\overrightarrow{E_{2\omega}}$  are the electric field amplitudes of the fundamental and second harmonic radiations.

## A Parallel polarizations of 400 nm and 800 nm fields

For the parallel polarizations,  $\overrightarrow{E_{\omega}} || \overrightarrow{E_{2\omega}}$ , the displacement of an electron, starting at  $t = t_0$  from the position  $r(t = t_0) = 0$  with zero velocity  $v(t = t_0) = 0$  is described by

$$r\left(t\right) = \frac{eE_{\omega}}{m\omega^{2}} \left(Sin\left(\omega t\right) - Sin\left(\omega t_{0}\right) - \omega\left(t - t_{0}\right)Cos\left(\omega t_{0}\right)\right) + \frac{eE_{2}\omega}{4m\omega^{2}} \left(Sin\left(2\omega t + \phi\right) - Sin\left(2\omega t_{0} + \phi\right) - 2\omega\left(t - t_{0}\right)Cos\left(\phi + 2\omega t_{0}\right)\right)\right)$$

$$(2)$$

The recombining electron returns to its parent ion at time  $t = t_1$ , such that  $r(t_1) = 0$ . The kinetic energy of this electron gained in the laser field is expressed as  $E_{kin} = m[v(t_1)]^2/2$ , where *m* is the mass of the electron, and  $v(t_1)$  is the final velocity of the electron. The



**Fig. 3.** Spectral density plot for different relative delays in the range of the delay times from -0.1 ps to +1 ps. A HWP was placed in the beam, so that the  $\omega$  and  $2\omega$  fields were parallel polarized; time step of 2.33 fs was used for the delay. HHs of even orders appear at delay ~0.8 ps, which corresponds to the temporal overlap of the red and blue pulses. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article).



**Fig. 4.** Comparison of HH spectra for orthogonal (blue solid line, showing high harmonic yields at delay time 0.5 ps from Fig. 2) and parallel (red dashed line, showing yields for delay time 0.8 ps from Fig. 3) polarizations of the overlapping 800 nm and 400 nm pulses. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article).



**Fig. 5.** The variations of the HHs yields with variations of the relative phase of  $E_{2\omega}$  and  $E_{\omega}$  fields: (a) The measured spectra of HHs; the phase difference  $\phi$  is incremented by  $\pi/4$  from bottom to top and the spectra are displaced for visual clarity. (b) The dependences of the yields of different order harmonics (14th -dashed red, 17th -solid green, 19th -dotted blue, 20th –dash-dotted purple) on the phase difference. $\phi$ .(For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article).

ponderomotive energy for the two-color case, defined as the average energy of an oscillating electron in the field is  $U_p = (e^2/m) [E_{\omega}^2/(4\omega^2) + E_{2\omega}^2/(16\omega^2)]$  [53]. The behavior of the electron in terms of the normalized ionization time ( $\tau_0 = \omega t_0/2\pi$ ), normalized recombination- time ( $\tau_1 = \omega t_1/2\pi$ ), which measure time in periods of the fundamental field, and its normalized kinetic energy ( $K_N$ ) expressed in  $U_p$  units at the moment of return to the parent ion is presented in Fig. 6 and Fig. 7. The intensity values are chosen as  $I_{\omega} = 3 \times 10^{14}$ W/cm<sup>2</sup> and  $I_{2\omega} \sim 7 \times 10^{13}$ W/cm<sup>2</sup> (similar to the experimental values), corresponding to the ratio  $E_{2\omega}/E_{\omega} \sim 0.5$  of the two-color field.

Fig. 6 shows the two-color electric field (*E*, grey dashed line) for the values of the relative phases  $\phi = 0$  and  $\phi = \pi/2$  and the kinetic energy of a returning electron (blue dash-dotted and orange solid lines) depending on the ionization-time of the electron. In the two-color field, the kinetic energy can exhibit several branches [54]. In Fig. 6(a) with  $\phi = 0$  one branch corresponds to the ionization time interval  $0.21 < \tau_0 < 0.42$  and the other branch to  $0.78 < \tau_0 < 0.98$ . At the field values indicated by green dots the maximum kinetic energy (shown by red dots) is attained. For  $\phi = \pi/2$  the two branches of the kinetic energy dependences correspond to the ionization time intervals  $0.25 < \tau_0 < 0.55$  and  $0.75 < \tau_0 < 0.92$ . For the branches with ionization at the negative phases of the field  $0.78 < \tau_0 < 0.98$  for Fig. 6(a) and  $0.75 < \tau_0 < 0.92$  for Fig. 6(b) the field reaches higher values, which should result in a higher ionization rate. Therefore, the contribution to the HHG from these second branches, shown in orange color can be expected to be predominant [19]. The kinetic energy of the returning electron and therefore the cutoff depend on the phase between  $\omega$  and  $2\omega$  fields as well as on the ionization time (Fig. 6(c)).

Depending on the ionization time long and short trajectories of the returning electron are possible. The long trajectory corresponds to earlier ionization and later recombination times, while the short trajectory corresponds to later ionization and earlier recombination times [53]. The long trajectory of the electron is related to a larger spread of the electron wave function, and therefore the probability of an electron going along this path to recombine is less, resulting in a reduced contribution to the HH radiation. Consequently, the electrons ionized in the region corresponding to short trajectory contribute predominantly to efficient HHG, especially when the



**Fig. 6.** Two branches of the solution for an electron ionized at the time shown on the horizontal axis and returning to the parent ion within one period of the fundamental field,  $(E_{2\omega}/E_{\omega})\sim 0.5$  and for the values of the phase difference  $\phi$ : (a)  $\phi = 0$ , (b)  $\phi = \pi/2$ . The kinetic energy is shown in units of  $U_p$ . Dash-dotted blue lines show the branches for the positive values of the field, and solid orange lines show the branches for the negative values of the field. Black dashed lines show the electric field with the scale in arbitrary units on the right-hand side. Red points label the maximum kinetic energies in each branch and the values of the field at the time of ionization are shown by respective green points. (c) The variation of the kinetic energy of the returning electron as a function of the normalized ionization-time ( $\tau_0$ ) and the normalized phase difference ( $\phi/(2\pi)$ ). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article).

variations of the harmonic phase are reduced [19]. The dipole created by electrons ionized in a single-color field is an odd function of time, changing the sign every half cycle of the field period, which results only in odd HHs. However, in a two-color field [54,55] the inverse symmetry of the system can be broken (as in Fig. 6(b)), which leads to the generation of even HHs as well. Also, in a two-color field, two branches of the solution for the returning electron are possible, which are shown in Fig. 7 for the experimental intensity parameters. For different values of the kinetic energy (horizontal axis) gained by the returning electron this figure shows the ionization and recombination times (on the vertical axes of the lower and upper figures) for two values of the relative phase (a)  $\phi = 0$ , and (b)  $\phi = \pi/2$ . The grey dotted line depicts the variations of the intensity of the two-color field with the scale in arbitrary units shown at the top of the figures. The arrows show the correspondence of the ionization and recombination for each of the solution branches, and the label (L) indicates long trajectories, while (S) shows short trajectories.

The kinetic energy of the returning electron varies depending on the ionization time and the phase difference  $\phi$ . For  $(E_{2\omega}/E_{\omega})\sim 0.5$ and the experimental values of the intensities of the fields, the maximum kinetic energy is  $E_{kin,max}=K_NU_p = 4.15U_p = 78.7$  eV, which is achieved for the normalized ionization time  $\tau_0 = 0.759$  and the phase difference  $\phi = 1.1\pi$ , corresponding to the cutoff of the HH spectrum at  $(E_i + Ekin, max)/h\omega \sim 61$  order (the ionization potential for Ar is  $E_i = 15.76$  eV).

#### B Perpendicular polarizations of 400 nm and 800 nm fields

Calculations of the kinetic energy of an electron acquired in a two-color field with perpendicular polarizations were also performed. The field is described by the same Eq. (1), where however  $\overrightarrow{E_{\omega}} \perp \overrightarrow{E_{2\omega}}$ . Consequently, the electron will be displaced in two perpendicular directions along  $\overrightarrow{E_{\omega}}$  and  $\overrightarrow{E_{2\omega}}$ , which we denote as  $r_{\omega}(t)$  and  $r_{2\omega}(t)$  These displacements (starting at  $t = t_0$  from the position  $r_{\omega}(t = t_0) = r_{2\omega}(t = t_0) = 0$  with zero velocity  $\nu_{\omega}(t = t_0) = \nu_{2\omega}(t = t_0) = 0$  are described by

$$r_{\omega}(t) = \frac{eE_{\omega}}{m\omega^2} (Sin(\omega t) - Sin(\omega t_0) - \omega(t - t_0)Cos(\omega t_0)),$$
(3)

$$r_{2\omega}\left(t\right) = \frac{eE_2\omega}{4m\omega^2} \left(Sin\left(2\omega t + \phi\right) - Sin\left(2\omega t_0 + \phi\right) - 2\omega\left(t - t_0\right)Cos\left(\phi + 2\omega t_0\right)\right)\right) \tag{4}$$

Again, we assumed experimental values for intensities and  $(E_{2\omega}/E_{\omega})\sim 0.5$ . For the electron ionized at time  $t_0$ , we can solve the equation  $r_{\omega}(t) = 0$  and find time  $t_1$ , when the electron returns to the initial position in this direction. However, for recombination the



**Fig. 7.** Ionization (lower plot) and recombination (upper plot) times corresponding to certain values of the kinetic energy normalized to  $U_p$  for  $(E_{2\omega}) \sim 0.5$  and two values of the phase difference of the  $\omega$  and  $2\omega$  fields: (a)  $\phi = 0$  (b)  $\phi = 0.5\pi$ . The total intensity of the field is shown as a dashed grey line. Blue (with higher maximum  $K_N$ ) and orange lines are the two solution branches for the positive and negative values of the field, respectively. Blue and orange arrows show the correspondence between the kinetic energy plots in terms of ionization and recombination times, L and S label long and short trajectories, respectively. For  $\phi = 0$  the ionization time intervals for long trajectories for the two branches are  $0.215 < \tau_0 < 0.255$  and  $0.786 < \tau_0 < 0.852$ , and the short trajectory intervals are  $0.255 < \tau_0 < 0.415$  and  $0.852 < \tau_0 < 0.975$ . For  $\phi = 0.5\pi$  the intervals for long trajectories are  $0.25 < \tau_0 < 0.354$  and  $0.774 < \tau_0 < 0.913$ . (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article).

electron must return to the parent atom simultaneously in both directions, thus enabling recombination, resulting also in the equation  $r_{2\omega}(t_1) = 0$ , which can be solved for  $\phi$ . Consequently, we obtain the unique relation between the ionization time  $(t_0)$ , phase difference  $(\phi)$ , and the normalized kinetic energy  $(K_N)$ .

The electron can return to the parent ion several times, and for each return, there is some probability of the recombination. The solutions for the electron to return (we calculated these solutions from 1st to 4th return) exist only in certain intervals of phase differences shown in Fig. 8, where  $\phi$  is normalized to  $2\pi$ . The changing phase difference  $\phi$  leads to changes of the return kinetic energy, and the following maxima are reached:  $E^{(1)}_{kin,max} = K_{N,1}U_p = 3.06U_p = 57.9 \text{ eV}$  at the absolute (unnormalized) phase value  $\phi = 0.17 \times 2\pi = 0.34\pi$  for the 1st return,  $E^{(2)}_{kin,max} = K_{N,2}U_p = 1.46U_p = 27.7 \text{ eV}$  at  $\phi = 0.47\pi$  for the 2nd return,  $E^{(3)}_{kin,max} = K_{N,3}U_p = 2.28U_p = 43.1 \text{ eV}$  at  $\phi = 0.46\pi$  for the 3rd return,  $E^{(4)}_{kin,max} = K_{N,4}U_p = 1.64U_p = 31.1 \text{ eV}$  at  $\phi = 0.48\pi$  for the 4th return. When the requirement for an electron to come back exactly to zero was imposed, only one solution with certain return kinetic energy was found for each phase difference and a fixed number of the electron return. All electron returns will contribute to the HHG, but their contributions are



**Fig. 8.** The kinetic energy of the returning electron: (a) the dependence of the maximum kinetic energy for each value of the normalized by 2  $\pi$  phase difference,  $\phi$  for the first (solid blue line), second (dotted black line), third (dashed orange line) and fourth (dash-dotted green line) electron returns; the solutions exist only within certain intervals of  $\phi$ . (b) the 3D plot of the kinetic energy for the first electron return on the normalized ionization time,  $\tau_0$  and the phase difference,  $\phi$ . Only one solution or no solution at all exist for each ( $\tau_0$ ,  $\phi$ ) pair and a given return number. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article).

expected to decrease for higher return numbers due to the larger spreading of the electron wavefunction. If we assume that the recollision happens when the distance of the electron from the origin is less than 4 a.u., which is the radius of the experimentally determined electron-argon impact cross-section [38,56], then for each value of the phase difference  $\phi$  there is a narrow interval of ionization times  $t_0$  with respective values of the electron kinetic energy (Fig. 9).

Fig. 10 shows calculation results for some specific phase differences for the case of perpendicular polarization for 1.5 periods of the  $E_{\omega}$  field (upper plots in figures (a–c)) and for the same time interval (3 periods of the  $E_{2\omega}$  field, lower plots). Depending on the phase difference a short trajectory ( $\phi = 0.05\pi$ , Fig. 10(a)), or a long trajectory ( $\phi = 0.49\pi$ , Fig. 10(c)) can be realized. Fig. 10(b) for  $\phi = 0.34\pi$  shows the trajectories of the electron in both directions returning with the maximum kinetic energy. For one period of the fundamental field the short trajectory intervals are  $0 < \phi < 0.34\pi$  and  $\pi < \phi < 1.34\pi$ , and the long trajectory intervals are  $0.34\pi < \phi < 0.5\pi$  and  $1.34\pi < \phi < 1.5\pi$ .

#### 4.1. Single-atom strong-field approximation (SFA) calculations

To get additional insights into the HHG process, we have used the Lewenstein's model [26] and its recent extension to a two-color excitation with circularly polarized laser beams [57] and calculated within the SFA approach the spectral components of the time-dependent dipole moment. Following the SFA assumption that in the transition of electrons from the ground state to the continuum the interaction with the intermediate excited states can be neglected and treating continuum states as plane waves, we obtain for the spectral components of the dipole moment

$$\vec{D}_{n} = \frac{1}{T} \int_{0}^{T} dt e^{i\omega t} \int_{-\infty}^{T} dt_{0} i e^{-iS_{pr}(t,t_{0})} \left[ \frac{2\pi}{i(t-t_{0})} \right]^{\frac{1}{2}} \times \vec{d}^{*} \left[ \vec{p}_{s}(t,t_{0}) - \vec{A}(t) \right] \left( \vec{d} \left[ \vec{p}_{s}(t,t_{0}) - \vec{A}(t_{0}) \right] \bullet \vec{E}(t_{0}) \right),$$
(5)

where atomic units are used,  $T = 2\pi/\omega$  is the period, *n* is the harmonic order, and  $S_{pr}(t,t_0) = \int_{t_0}^{t} dt' \left\{ \left[ \vec{p}_s(t',t_0) - \vec{A}(t') \right]^2 / 2 + E_i \right\}$  is

action corresponding to stationary phase,  $\vec{A}(t) = \int dt' \vec{E}^t(t')$  is the vector potential, which is related to the electric field,  $\vec{p}_s(t,t_0) = \int_{t_0}^{t_0} dt' \vec{A}(t')/(t-t_0)$  is the stationary momentum and we use the following dipole matrix approximation [26]:  $\vec{d}(\vec{p}) = \int_{t_0}^{t_0} dt' \vec{A}(t')/(t-t_0)$ 

 $dt' \vec{A}(t')/(t-t_0)$  is the stationary momentum and we use the following dipole matrix approximation [26]:  $\vec{d}(\vec{p}) = i(2^{7/2}\alpha^{5/4}/J_1)\vec{p}/(\vec{p}^2+\alpha)^3$ ,  $\alpha = 2E_i$ ,  $E_i$  is the ionization potential.

Approximating the integrals of the quickly oscillating function with the stationary-phase method, we get two equations for finding the stationary points:

$$\frac{1}{2}[p_s(t,t_0) - A(t_0)]^2 = -E_i, n\omega = \frac{1}{2}[p_s(t,t_0) - A(t_0)]^2 + E_i.$$
(6)

The yields of high harmonics are proportional to the squares of the dipole moment spectral components that were found for a singleelectron recombination event:  $I_n \propto \left| \vec{D}_n \right|^2$ , so the dependence  $\left| \vec{D}_n(n) \right|^2$  presents the dipole spectral intensity.

The dipole spectral intensity for parallel polarizations for high harmonics is shown in Fig. 11. The calculations were performed for phase differences  $\phi = 0$  and  $\phi = \pi/2$ . According to Fig. 7 both solution branches contribute to harmonics of up to ~42 order and for harmonics with order exceeding this number only first branch contributes. For each harmonic there are four (for n<42) and two (for n>42) pairs of ionization and recombination times following from classical equations Eq. (1,2), which were used as starting values for



**Fig. 9.** The normalized kinetic energy  $K_N$  of a returning electron at different phase differences  $(\phi/2\pi)$  in the assumption that the re-collision happens, if its distance from the origin is less than 4a.u. For each value of  $\phi$  there can be some interval of values of  $K_N$ , related to solutions with different ionization times, or no solutions at all. The smaller arc within the larger one in the figure corresponds to the second return of the electron. Compared to Fig. 8 the gap with no solutions becomes narrower.



**Fig. 10.** Calculations of electron trajectories for some specific phase differences for perpendicular case: (a)  $\phi = 0.05\pi$  (short trajectory) (b)  $\phi = 0.34\pi$  (maximum kinetic energy), (c)  $\phi = 0.49\pi$  (long trajectory). In (a-c) the upper figures are the solutions for the plane with the fundamental field, the lower figures are the solutions for the plane with the second harmonic field; red dashed lines show the return time for the maximum kinetic energy, which separates the return times for short and long trajectories,  $t_N = \omega t/2\pi$  is normalized time. Black dashed lines are the electric fields with the scale in arbitrary units on the right-hand side. Blue lines are the electron trajectories for the  $E_{\omega}$  and  $E_{2\omega}$  fields in their respective planes. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article).



**Fig. 11.** Dipole spectral intensity for dual-color excitation with parallel polarizations calculated for different phase delays:  $\phi = 0$  (black circles) and  $\phi = 0.5\pi$  (blue squares). The thin solid lines are the guides for the eye. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article).

finding complex solutions of Eq. (6). Consequently, the cutoff of the second branch exhibits a stepwise decline of the intensity with significant intensity variations (Fig. 11). The interference of the contributions of the two branches with long and short trajectories leads to oscillations of the yield of high harmonics, which is especially pronounced near the cutoff regions for the solution branches.

The dipole spectral intensity for perpendicular polarizations is shown in Fig. 12. The calculations are performed for phase differences  $\phi = 0.34\pi$  (this corresponds to the highest kinetic energy according to Fig. 8,9,  $E_{kin} = 57.9$  eV, cutoff n = 47),  $\phi = 0.4\pi$  ( $E_{kin} = 49$  eV, cutoff n = 42) and  $\phi = 0.2\pi$ . The indicated cutoffs are in agreement with the spectra of Fig. 12 obtained with the SFA quantum-mechanical approach.

#### 5. Discussion

Our experiments demonstrate that the generation of even harmonics takes place only when there is a combination of the  $\omega$  and  $2\omega$  fields. The experimental and calculational results show that the HHG process in a two-color field for parallel and perpendicular polarizations is different. The comparison of the spectral densities for orthogonal and parallel polarizations (Fig. 4) shows that the yield of the XUV radiation is higher for the orthogonal case. The highest harmonic yields for odd harmonics were observed near-zero delays. The yields of HHs from 13th to 21st orders for orthogonal polarizations are higher compared to the parallel polarization by a factor ranging from 2 to 9 (Fig. 4). A possible explanation is the following. In the parallel case, the emission of radiation by different electrons is not synchronized, since there is a broad interval for the returning electrons, which results in radiation emission with different phases. Consequently, the interference of all these contributions with various phases leads to a relatively low output. For perpendicular polarizations, the variations of the phases of partial contributions to generated HHs are much smaller, since the phases of the driving fields vary only slightly across these beams, and the first return of electrons happens almost simultaneously in the whole cross-section, independently of the local amplitude of the electric field, which results in emissions with close phases. This synchronization of the



**Fig. 12.** Dipole spectral intensity for dual-color excitation with perpendicular polarizations calculated for different phase delays:  $\phi = 0.34\pi$  (black circles: corresponds to the maximum kinetic energy of the returning electron, see Fig. 8(a)),  $\phi = 0.2\pi$  (blue squares) and  $\phi = 0.4\pi$  (brown diamonds). The thin solid lines are the guides for the eye. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article).

electron returns has important implications for the generation of attosecond pulses, since the generation of much shorter pulses can be expected in this case, which requires further investigation. Besides such a synchronization another factor should be taken into account. With the high-intensity two-color field, such as we apply, the degree of ionization can also affect the yield of HHs. At the conditions of our experiment, the parallel polarized two-color field provides a somewhat higher peak intensity and also higher ionization, which can lead to an additional phase mismatch. For the used intensities the Keldysh parameter [58],  $\gamma = (I_p/2U_p)^{0.5} \sim 0.65 < 1$ , which indicates that the tunneling ionization prevails and the ADK theory [59] is applicable. Calculating the ionization fraction  $\xi$  of Ar after the passing of the two-color field with experimental values of intensities and  $\phi = 0$  gives  $\xi = 35\%$  for perpendicular polarizations. Furthermore, the higher electron density can result in higher outward divergence of the radiation [20]. These factors can account for significantly higher yields of HHs for the case with  $E_{ap}$  and  $E_{2ap}$  perpendicular polarizations.

The calculations for perpendicular polarizations also show that for each value of the phase difference of the  $E_{\omega}$  and  $E_{2\omega}$  fields and the first return of the electron (the contributions of higher-order returns should be significantly smaller) there is only a narrow interval of the electron return times, corresponding to solutions with the return kinetic energies also within a relatively narrow range (especially at higher energies). Consequently, one can expect that by changing the phase difference it is possible to control the kinetic energy, and thus also the cutoff energy of the emitted photons, resulting from the recombination. In the experiment, the beams have a distribution of intensities over their cross-section, which results in a broadened distribution of kinetic energies of returning electrons. Then as it follows from the semi-classical viewpoint, the spectrum of HHs should contain all harmonics up to the highest, which corresponds to the maximum kinetic energy for a given value of the phase difference. Figs. 8 and 9 show a strong dependence of the kinetic energy on the phase difference; therefore, variations of the latter should lead to variations of the harmonic yields, especially for the highest harmonics.

The experimental results (Fig. 5) show that indeed the variations of the phase difference lead to changes of the harmonics yields, and the highest harmonics show the largest relative yield variations. For instance, as it follows from Fig. 5(b) 14H and 17H show variations ~200 %, 19H varies by ~300 %, and 20H exhibits variations up to ~500 %. The fact that the yield does not disappear for certain intervals of the phase difference, as it follows from the results of Figs. 8 and 9, can be related to two circumstances. First, the recombination of the electron can happen even if it gets within the radius of the characteristic electron-argon impact cross-section of ~4 a.u. [38,56]. Second, according to the semi-classical interpretation, the macroscopic radiation of certain high harmonic registered in the experiment is produced in the whole volume with the cut-off below the energy of this high harmonic, and as a result, volumetric averaging takes place [60]. However, such volumetric averaging is reduced for the most energetic photons, corresponding to the highest intensity in the center of the laser beams, and consequently, larger relative oscillations for the highest harmonics can be expected, as was observed in the experiment (Fig. 5).

The calculations of Fig. 10 demonstrate that by varying the phase difference between  $E_{2\omega}$  and  $E_{\omega}$  fields it is possible to select which trajectory, long or short will be realized for the returning electron, which was confirmed experimentally [38]. Also switching between dominating short or long trajectory enables control of the HH divergence [33].

The calculations with the SFA model show that the HHG by a two-color field with parallel and perpendicular polarizations exhibits different harmonic spectra, which in both cases depend on the relative phase of the  $\omega$  (IR) and  $2\omega$  (SH) fields. For the case of perpendicular polarizations, the harmonic intensity and cutoff vary with the changing relative phase, which corroborates the experimental observations of Fig. 5. To accurately account for the yields of high harmonics, macroscopic phenomena involving ionization saturation effect and phase matching should be taken into account.

#### 6. Conclusions

We studied high order harmonic generation by a combination of laser pulses centered at two wavelengths at 800 nm and 400 nm. The variation of the relative delay and the overlapping of these infrared (IR) and the second harmonic (SH) fields can lead to the

generation of even order HHs as well as to the enhancement of the odd harmonics. The relative delay between the two fields has a strong influence on the generated harmonic spectra. When this delay is changed for the overlapping pulses, the HH yields experience oscillations. Such an effect was most pronounced for perpendicular polarizations of the IR and SH fields and the highest harmonic orders. This was discussed in relation to electron behavior in a two-color field that revealed significant differences for the parallel and perpendicular cases. Within one period of the fundamental field, for the former, there exists a broad time interval for returning electrons, and in the latter case there are only two narrow intervals for possible solutions for each of the electron returns to the parent ion for a fixed phase difference between the  $\omega$  and  $2\omega$  fields. This synchronization of much shorter pulses can be expected in this case. The calculations of the harmonic intensities within the strong field dipole approximation were also performed. The presented additional possibilities of controlling the yields of high harmonics and the content of the XUV spectra by using two-color fields can find applications in the ultrafast absorption spectroscopy, imaging, studies of biological structures, and materials science.

#### Disclosures

The authors declare no conflicts of interest.

### **Declaration of Competing Interest**

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper

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