Journal of Physics B: Atomic, Molecular and Optical Physics

Theoretical study of single attosecond pulse generation with a three-colour laser field

J. Phys. B: At. Mol. Opt. Phys. **42** (2009) 225601 (5pp) doi:10.1088/0953-4075/42/22/225601

Theoretical study of single attosecond pulse generation with a three-colour laser field

Rui-Feng Lu, Hai-Xiang He, Ya-Hui Guo and Ke-Li Han

State Key Laboratory of Molecular Reaction Dynamics, Dalian Institute of Chemical Physics, Chinese Academy of Sciences, Dalian 116023, People's Republic of China

E-mail: klhan@dicp.ac.cn

Received 21 July 2009, in final form 16 September 2009
Published 5 November 2009
Online at stacks.iop.org/JPhysB/42/225601

Abstract

We present a method of producing single attosecond pulses by using a few-cycle (5 fs) driving pulse with two additional weak control pulses. We discuss how single attosecond pulses produced from high-order harmonic generation processes in a synthesized three-colour laser field are similar to those processes in a much shorter single-colour laser field. Based on the high-order harmonic spectrum, classical ionizing and returning energy maps, time–frequency maps and time profiles of the attosecond pulses, the actions of the synthesized three-colour laser field are analogous to a 3 fs field although some differences still exist, and our method is proved to be a potential way to reduce the attosecond pulse duration from high-order harmonic generation with a currently available ultrafast laser source instead of a shorter pulse.

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

1. Introduction

Attosecond extreme ultraviolet (XUV) pulses open the way to a new regime of investigating and manipulating basic ultrafast electronic processes with unprecedented precision [1], and the progress in attosecond science and technology has been reviewed recently [2–7]. An isolated attosecond pulse is preferred for detecting and controlling the electronic dynamics, thus followed the generation of a train of attosecond pulses with a multicycle laser [8], a single isolated XUV pulse with the duration of 130 as has been lately generated by the polarization gating technique [9]. To date, high-harmonic generation (HHG) is the most promising method for producing attosecond pulses. To further shorten the XUV pulse duration, a straightforward approach is to employ intense sub-1.5-cycle (3.3 fs at 720 nm) optical pulses in rare gases [10], because from a theoretical point of view, the shorter the driving pulse, the broader the attainable XUV supercontinuum in the HHG spectrum [11]. The short pulse (<5 fs), however, still presents technical challenges in experimental conditions.

A practical way of creating isolated XUV pulses is to use a two-colour laser field synthesized by a low-intensity second-harmonic beam with the fundamental beam to control the HHG process. Many previous theoretical and experimental studies have confirmed that the XUV supercontinuum generation is a highly nonlinear optical process sensitive to a slight change in a dichromatic laser field [12–15]. More recently, a dubbed double optical gating approach, with dependence of the HHG on 2-periodicity carrier-envelop phase, was demonstrated to efficiently control and generate single attosecond pulse [16]. According to the classical 'three-step' model [17], the significantly varying electric field amplitude from one half-cycle to the next, and the difference of maximum kinetic energy of an electron returning to its parent ion for each half-cycle, are responsible for the XUV supercontinuum. In spite of some technical difficulties in the experiment like the requirement of state-of-the-art few-cycle main driving pulses, an interesting question emerges in regard to what would happen if multicolour fields were used. That concept involves optimal control of laser fields, which is much more complicated to accomplish in multicolour fields than in two-colour fields. It is well known that attosecond pulses can also be generated from coherent Fourier synthesis of independent multicolour sources [18]. Keeping in mind that idea and the principle of HHG, we theoretically synthesize a three-colour laser field with few-cycle lasers, and interact with helium (He)

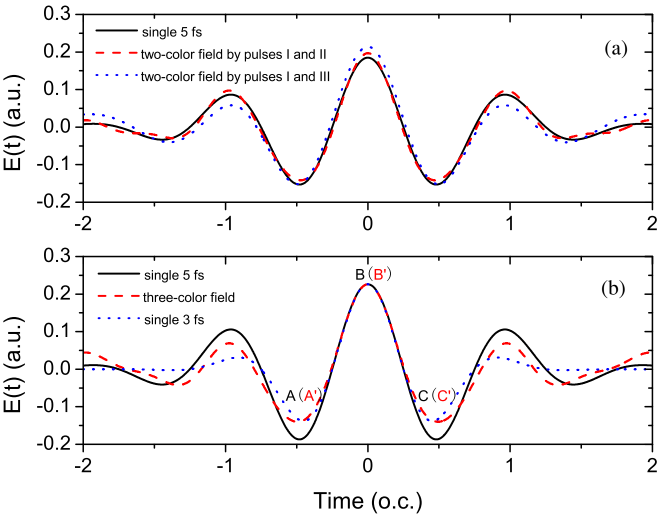

Figure 1. (a) Electric fields of the 5 fs/800 nm pulse (I), the two-colour pulse synthesized from pulse I and 25 fs/400 nm pulse (II), and the two-colour pulse synthesized from pulse I and 25 fs/1600 nm pulse (III). (b) Electric fields of single 3 fs and 5 fs pulses (800 nm), and the three-colour pulse synthesized from pulses I, II and III.

to elucidate the role of the synthesized field for attosecond pulse generation.

2. Theory and discussion

In the present quantum wavepacket simulation, a 5 fs/800 nm pulse (pulse I), a 25 fs/400 nm pulse (pulse II) and a 25 fs/1600 nm pulse (pulse III) are combined to serve as the driving pulse. The intensities are chosen to be 1.2×10^{15} W cm^{-2}, 5×10^{12} W cm^{-2}, and 3×10^{13} W cm^{-2}, respectively. The synthesized field can be expressed as

$$E(t) = E_1 f_1(t)\cos(\omega t) + E_2 f_2(t)\cos(2\omega t) + E_3 f_3(t)\cos(0.5\omega t), \quad (1)$$

where ω is the frequency for the 800 nm fundamental pulse, and E_1, E_2 and E_3 are the electric field amplitudes of the fundamental pulse, the second-harmonic pulse and the subharmonic control pulse, respectively. The other two fields can be generated by optical processes from the fundamental 800 nm laser field. The Gaussian pulse envelope is chosen, with the formula

$$f_i(t) = \mathrm{e}^{-2\ln(2)t^2/2\tau_i^2}, \qquad i = 1, 2, 3 \ldots, \quad (2)$$

where τ_i is the full width at half maximum (FWHM).

The dashed line in figure 1(a) shows the electric field synthesized by the main pulse I and control pulse II. It is clear that the positive amplitude of the main peak is enhanced from a constructive interference, and in contrast, the negative peak amplitudes of the two adjacent half-cycles are suppressed because of a destructive interference. The dotted line shown in figure 1(a) is the electric field synthesized by the main pulse I and control pulse III. Note that the second strongest positive peak amplitudes are suppressed, whereas the negative peak amplitudes neighbouring the main peak remain invariable. Utilizing these characteristics of the control pulses, the role of the three-colour field seems to be essentially the shortening of the pulse duration, as seen in figure 1(b). The main similarity between the three-colour field and 3 fs is the FWHM (not depicted here with the envelop profile). It is a smart scheme, albeit a little bit simple and intuitive which is still that one be in the few-cycle regime and use phase-stabilized pulses. Note that the laser peak intensities of the single 3 fs and 5 fs pulses equal that of the synthesized three-colour pulse, specifically, $I = 1.794 \times 10^{15}$ W cm^{-2}, corresponding to a ponderomotive energy $U_p = E^2/4\omega^2$ of 107 eV. Here, we establish an ultimate goal that the single attosecond pulse generation from the HHG in the 3 fs laser pulse can be obtained through the 5 fs pulse, which is the shortest laser available with present experimental conditions. In fact, two-colour fields can also achieve a similar effect [19]. Nevertheless, our optimal calculations indicate that it is difficult to reach the goal as well as the three-colour field. Moreover, in order to keep the synthesized electric field symmetric, as with a shorter 3 fs laser field, it is not necessary to alter the relative phase shifts among the laser pulses I, II and III; hence, the optimal adjustments have been simplified to a certain extent.

To investigate the HHG spectrum and the attosecond pulse generation, we numerically solve the time-dependent Schrödinger equation (TDSE) by means of the sine discrete variable representation (DVR) and split-operator method. The method has previously been applied to study nonadiabatic quantum scattering dynamics [20] and to track nuclear and electron wavepackets in a strong laser field [21, 22]. For our simulation, a soft-core potential $V(x) = -1/\sqrt{\alpha + x^2}$ with the parameter α of 0.484 was chosen, so that the ionization energy I_p of 24.6 eV for the ground state of He is close to the experimental value. The harmonic spectra are obtained by Fourier transforming the time-dependent dipole acceleration from the Ehrenfest theorem [23], and the ultrashort attosecond pulses are generated through superposing several harmonics (i.e., inverse Fourier transformation). All the calculations were performed with our parallel quantum wavepacket computer code LZH-DICP [24].

Figure 2 depicts the HHG power spectra of atomic He for the three-colour laser field as well as for 3 fs, 4 fs and 5 fs laser fields with the same peak intensities as that of the three-colour laser field. The differences of total amplitudes among the four curves in the plateau region are actually small, so we shifted three curves for the purpose of clarity so that the harmonic signals of 3 fs and 4 fs pulses, and three-colour field are scaled. The spectra clearly display the structure as expected: it is irregular for the low harmonics and fascinatingly becomes regular and continuous for the high harmonics. Attention should be paid to the differences below the harmonic cutoff because they are the ones which come to dominate the experimental signal if not carefully filtered out. As the length of the pulse decreases, the HHG continua in the cutoff region become broader, and even two plateaus can be observed for 3 fs laser. Regarding the spectrum in the combination of the 5 fs driving pulse and two 25 fs control pulses, a pattern very similar to that of 3 fs laser is found: there are two plateaus and a broad continuum with a bandwidth of approximately 195 eV in the second cutoff regime (compared with 215 eV for the

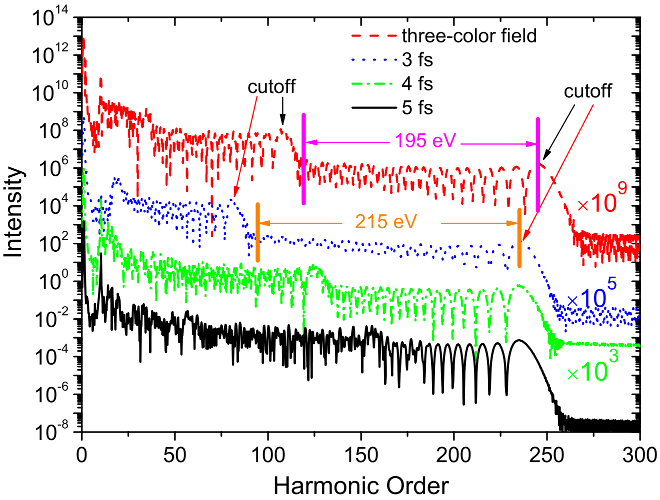

Figure 2. HHG power spectra in single 3 fs, 4 fs and 5 fs fields as well as in the three-colour field. For the purpose of clarity, the harmonic intensities of 4 fs and 3 fs pulses and three-colour field are multiplied by factors of 10^3, 10^5 and 10^9, respectively.

3 fs laser). From the illustration of figure 1, it is confirmed that the different cutoffs in this figure were also observed in a recent experiment as so-called half-cycle cutoffs [25].

HHG can qualitatively be explained by the classical model [17], with figures 3(a) and (b) presenting the dependence of the kinetic energies E_k on the ionization and recombination times for the 3 fs laser field and the three-colour field, respectively. In figure 3(a), we focus on peaks that are labelled as A, B and C (see also figure 1(b)). At point A, the electron ionizing in the previous half-cycle returns to the parent ion core, gaining maximum kinetic energy. Also, the electron tunnel ionizes when the laser field reaches the local maximum at point A, then gains energy from the following peak field B and recombines to the parent ion core between B and C; similarly, the electron ionizing from B will be accelerated by C and return after C. Therefore, peak A contributes to the harmonics from the 67th order to the 81th order, and only peak B contributes to the harmonics higher than the 95th order. In terms of the three-colour field (figure 3(b)), in the same way peak A′ supports the harmonics between the 87th and 107th orders, and only peak B′ supports the harmonics higher than the 119th order.

In order to better understand the physics behind the broadened XUV continuum spectra with two cutoffs in the 3 fs and three-colour lights, wavelet time–frequency analyses [11] have been performed, as presented in figures 3(c) and (d) which present more information such as amplitudes. It can be seen that three main peaks contribute to the harmonics both in figure 3(c) (peaks A, B and C) and figure 3(d) (peaks A′, B′ and C′). The first plateau for 3 fs (three-colour field) results from the harmonics between peaks A (A′) and C (C′), whereas the second plateau for 3 fs (three-colour field) originates from the harmonics between peaks A (A′) and B (B′). The difference between A (A′) and B (B′) is 215 eV (195 eV), respectively, which is consistent with the results in figure 2 as well as in figures 3(a) and (b). It can be seen from amplitudes that there is a very strong presence of the long trajectory in figure 3(d) of the three-colour field and it is the reason why the magnitudes of high-order harmonics in the cutoff region fluctuate severely in the case of the three-colour field in figure 2. Although there are some differences between these two pulses in figure 3(c) and (d), the classical and quantum pictures provide consistent

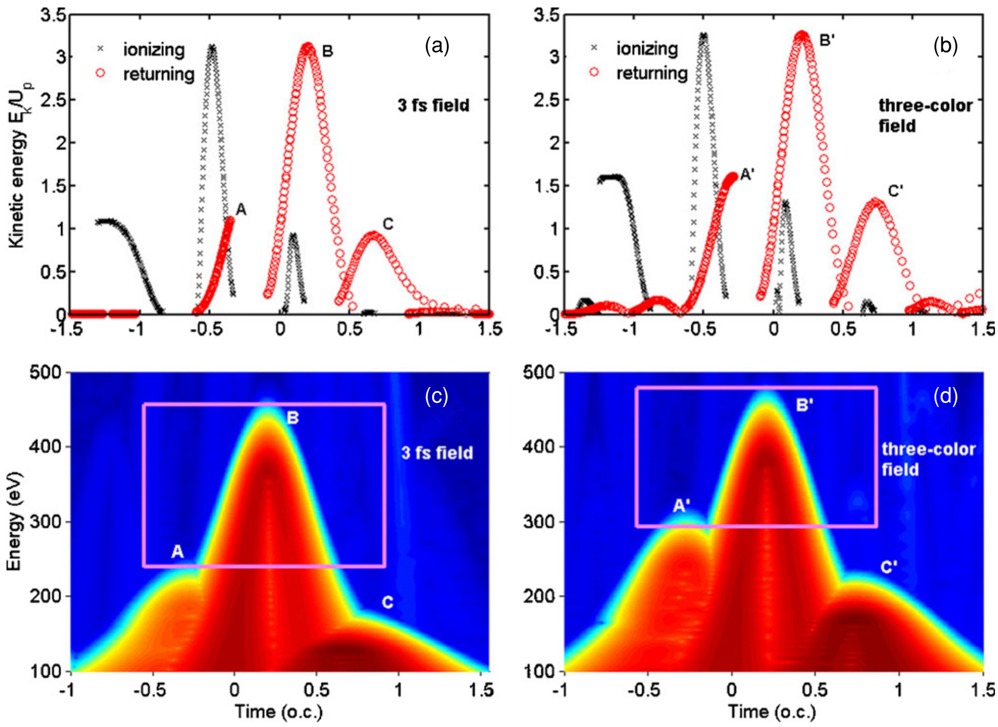

Figure 3. Classical ionizing and returning energy maps and wavelet time–frequency profiles of the HHG spectra in (a), (c): 3 fs field, and (b), (d): three-colour field. Note that the colour scale in (c) and (d) is in logarithmic units.

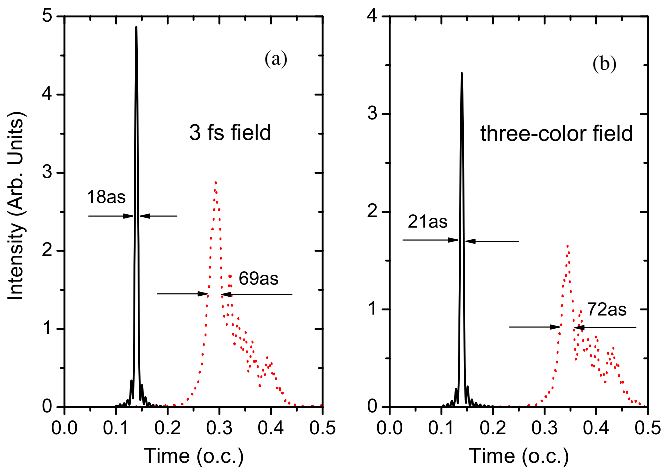

Figure 4. The temporal profiles of the attosecond pulses generated from the XUV continua without phase compensation, the results with phase compensation are also shown as solid lines: (a) single 3 fs field, (b) three-colour field.

and complementary information regarding the mechanism for the HHG and the creation of attosecond XUV. The harmonics higher than the 95th order in the 3 fs laser field and the 119th order in the three-colour field can be filtered out to generate attosecond pulses. Next, in figure 4, we show the temporal profile of the attosecond pulse by simple inverse Fourier transformation of spectral continuum with a bandwidth of 215 eV for the 3 fs laser field and the 195 eV for the three-colour field, thereby giving out isolated 69 as and 72 as pulses without phase compensation. By compensating for the chirp of XUV continuum which is on the assumption that the phases of harmonics are the same, an isolated 18 as pulse close to the Fourier transform limit with a clean profile can be attained in the 3 fs field, with 21 as attained for the three-colour field. Although the overall amplitudes of harmonics in the cutoff region between these two laser fields are almost the same, there are differences for both single attosecond pulses synthesized from the quite different continua, and further due to the presence of the long trajectories in the three-colour field. It should be restated that we aimed to employ available experimental lasers to produce single attosecond pulses that could be generated for a shorter driving laser pulse. That goal has been achieved by employing a synthesized three-colour field. It is noteworthy that single few-cycle or multicycle pulses ($\geqslant$5 fs) do not generate such broad continuum in high-order harmonics, and short attosecond pulses as those generated by the above-mentioned single 3 fs pulse and synthesized three-colour field.

3. Summary

To summarize, we investigated the role of a three-colour laser field for the generation of a single attosecond pulse in the ultrabroad XUV continuum. Through detailed comparisons with single 3 fs laser field and analyses, we exposed the underlying physics of a strong fundamental driver pulse with additional weak control pulses. In the three-colour laser field, the sub-100 as pulses generated from the broad bandwidth of 195 eV without interference by the neighbouring half-cycles, and the temporal resolution even allowed us to approach subatomic time unit by compensating the chirp with proper media which are similar to the results of a single 3 fs laser field although with some differences. With the ongoing research, the selection of a specific short or long quantum trajectory in the few-cycle regime attracts much attention [26, 27], and interestingly, HHG can efficiently be enhanced in an orthogonally polarized two-colour laser field [28]. These viewpoints should be applied to a synthesized three-colour field with adjustment of intensity ratio and time delay among the three pulses. It is hopeful that with a multicolour field one could use this method to reduce the pulse duration producing single attosecond pulses from HHG, especially the long-duration laser field and alleviate the reliance on short pulses. Further research will likely demonstrate that continued refinement of three-colour or multicolour schemes coupled with advances in experimental technology will allow the ability to produce elegant single attosecond pulses.

Acknowledgments

This work was supported by the knowledge innovation program of the Chinese Academy of Sciences (DICP K2007F2).

References

[1] Drescher M, Hentschel M, Kienberger R, Uiberacker M, Yakovlev V, Scrinzi A, Westerwalbesloh T, Kleineberg U, Heinzmann U and Krausz F 2002 *Nature* **419** 803–7
[2] Agostini P and Dimauro L F 2004 *Rep. Prog. Phys.* **67** 813–55
[3] Corkum P B and Krausz F 2007 *Nat. Phys.* **3** 381–7
[4] Osborne I and Yeston J 2007 *Science* **317** 765–6
[5] Bucksbaum P H 2007 *Science* **317** 766–9
[6] Goulielmakis E, Yakovlev V S, Cavalieri A L, Uiberacker M, Peryak V, Apolonski A, Kienberger R, Kleineberg U and Krausz F 2007 *Science* **317** 769–75
[7] Kapteyn H, Cohen O, Christov I and Murnane M 2007 *Science* **317** 775–8
[8] Paul P M, Toma E S, Breger P, Mullot G, Augé F, Balcou P, Muller H G and Agostini P 2007 *Science* **292** 1689–92
[9] Sansone G *et al* 2006 *Science* **314** 443–6
[10] Goulielmakis E *et al* 2008 *Science* **320** 1614–7
[11] Carrera J J, Tong X M and Chu Shih I 2006 *Phys. Rev.* A **74** 023404
[12] Dudovich N, Smirnova O, Levesque J, Mairesse Y, Ivanov M Y, Villeneuve D M and Corkum P B 2006 *Nat. Phys.* **2** 781–6
[13] Pfeifer T, Gallmann L, Abel M J, Neumark D M and Leone S R 2006 *Opt. Lett.* **31** 975–7
[14] Mauritsson J, Johnsson P, Gustafsson E, L'Huillier A, Schafer K J and Gaarde M B 2006 *Phys. Rev. Lett.* **97** 013001
[15] Zeng Z N, Cheng Y, Song X H, Li R X and Xu Z Z 2007 *Phys. Rev. Lett.* **98** 203901
[16] Mashiko H, Gilbertson S, Li C, Khan S D, Shakya M M, Moon E and Chang Z 2008 *Phys. Rev. Lett.* **100** 103906
[17] Corkum P B 1993 *Phys. Rev. Lett.* **71** 1994
[18] Yoshitomi D, Kobayashi Y, Takada H, Kakehata M and Torizuka K 2005 *Opt. Lett.* **30** 1408–10
[19] Pfeifer T, Gallmann L, Abel M J, Nagel P M, Neumark D M and Leone S R 2006 *Phys. Rev. Lett.* **97** 163901

[20] Chu T S, Zhang Y and Han K L 2006 *Int. Rev. Phys. Chem.* **25** 201–35
[21] Hu J, Han K L and He G Z 2005 *Phys. Rev. Lett.* **95** 123001
[22] Hu J, Wang M S, Han K L and He G Z 2006 *Phys. Rev.* A **74** 063417
[23] Burnett K, Reed V C, Cooper J and Knight P L 1992 *Phys. Rev.* A **45** 3347
[24] Lu R F, Zhang P Y and Han K L 2008 *Phys. Rev.* E **77** 066701
[25] Haworth C A, Chipperfield L E, Robinson J S, Knight P L, Marangos J P and Tisch J W G 2007 *Nat. Phys.* **3** 52–7
[26] Schafer K J, Gaarde M B, Heinrich A, Biegert J and Keller U 2004 *Phys. Rev. Lett.* **92** 023003
[27] Song X, Zeng Z, Fu Y, Cai B, Li R, Cheng Y and Xu Z 2007 *Phys. Rev.* A **76** 043830
[28] Kim I J, Kim C M, Kim H T, Lee G H, Lee Y S, Park J Y, Cho D J and Nam C H 2005 *Phys. Rev. Lett.* **94** 243901