Two-color field for the generation of an isolated attosecond pulse in water-window region

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Abstract: For the investigation of various ultrafast electron dynamics, an isolated attosecond pulse in a broad spectral range is necessary. The generation of isolated attosecond pulses demands the manipulation of the electric field of a laser. We propose a two-color field scheme for generating an isolated attosecond pulse in the water-window region. Two-color fields are generated by mixing two equally-strong pulsed color fields. The investigation shows that an isolated attosecond pulse with a photon energy of near 500 eV and a pulse duration of 125 – 188 attoseconds can be generated using 10 - 15 fs FWHM laser fields.

OCIS codes: (320.0320) Ultrafast optics; (190.4160) Multiharmonic generation; (190.2620) Harmonic generation and mixing.

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1. Introduction

The attosecond science is a newly emerging area, which concerns about electron dynamics at attosecond time scale and deals with the development of new methodology related to this investigation. To measure and control electron motion in its own natural time scale is necessary to further understand matter on as-yet unprecedented time scale and manipulate matter in totally new ways. Hence an isolated attosecond pulse is prerequisite to attosecond science. To generate an isolated attosecond pulse, several methods have been developed such as amplitude gating by < 4 fs laser pulses [1,2], polarization gating [3], double optical gating (DOG) [4], ionization gating [5], generalized double optical gating(GDOG) [6] and two color mixing [7], based on high-order harmonics generation (HHG).

These isolated attosecond pulses have been used in studying electron dynamics in atoms [8–10], in molecules and in condensed matter system [11]. These are remarkable achievements; yet, for wider spread of attosecond pulses in various electron dynamics, attosecond pulses of different photon energies still need to be produced.

The effect of the mixing of pulsed two color fields on the generation of an isolated attosecond pulse has been investigated. Recently, a continuum around ~80 eV was obtained by a superposition of fields at 1290nm and 780nm [12]. Moreover, a continuum around ~200 eV was generated by an intense ultrashort two-color driver [13]. It was also reported that the difference between the square of the strongest two-color field amplitude and the 2nd strongest two-color field amplitude is approximately proportional to the continuum length, thus inversely proportional to the attosecond pulse duration [7]. In other words, the bigger the difference is, the narrower the isolated attosecond pulse will be.

Theoretically, a two-color field generated by mixing two equally strong laser fields can favorably have a larger difference between its strongest amplitude and the next strongest one, if the wavelength of the component colors is properly chosen. Recent years have observed the advent of femtosecond, $1 - 2$ µm light pulse with a pulse energy of a few mJ via optical parametric amplification (OPA) method [14]. These sources were used to generate HHG [15]. It is now necessary and worthwhile to do systematic works to find out what kind of isolated attosecond pulses a two-color field of equally-strong visible and IR field can generate.

This paper reports that the proper mixing of two equally strong laser fields can lead to isolated attosecond pulses as short as 125 as inside the water window region. This extension of the photon energy allows us to investigate the quantum effects in atoms and molecules on a broader photon energy range [16].

2. Continuum analysis

To facilitate discussion, first, we define the ratio R of the two-color field as $R = E_{1st} / E_{2nd}$. E_{1st} denotes the strongest amplitude in the two-color field, while E_{2nd} denotes the second

strongest one. The intensity difference ratio is given by $\delta_d = \dfrac{E_{1st}{}^2 - E_{2nd}{}^2}{E_{1st}{}^2}$. Generally speaking, the larger the ratio R is, the larger the intensity difference ratio δ_d is, which results in a longer continuum length and thereby a shorter attosecond pulse duration. The estimation of this ratio leads us to determine what kinds of colors are used as the components of the two-color field in our simulations. Considering the real experiment conditions, we fix the intensity of these two laser fields at 3×10^{14} W/cm^2, which is readily available and strong enough for HHG. Moreover, since 800 nm light is the most popular wavelength of fs lasers, we fix the wavelength of one of the two colors at 800 nm. To determine the wavelength of the second laser field, we need to consider two facts: first, longer driving wavelengths are expected to extend the HHG cutoff since the maximum photon energy scales as the square of the driving field wavelength [17,18]; on the other hand, by taking the carrier wavelength longer, the electron wave packet experiences a longer transit time before recolliding and thus a higher quantum-mechanical diffusion, which leads to a decrease of the HHG process efficiency [19]. Therefore, the wavelength ranging from 300 nm to 2800 nm seems to be a good compromise between these two conflicting requirements. The pulse duration of both color fields is placed between 5 and 15 fs full-width-at-half-maximum (FWHM).

To find an optimum wavelength of the 2nd color, we have done a series of simulations for various wavelengths of the 2nd color from 300 nm to 2800 nm for a fixed pulse duration to find out the highest value for R. The results are shown in Fig. 1.
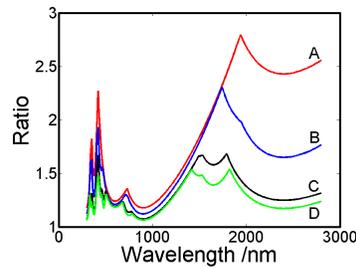

Fig. 1. The change of the ratio, R, with respect to the wavelength of the second color for a given pulse duration. The wavelength of the first color is fixed at 800 nm. Different color corresponds to different pulse durations. A (Red): 5 fs FWHM for 800 nm, 5 fs FWHM for 2nd color; B (blue): 5 fs FWHM for 800 nm, 10 fs FWHM for 2nd color; C (black): 10 fs FWHM for 800 nm, 10 fs FWHM for 2nd color; D (green): 10 fs FWHM for 800 nm, 15 fs FWHM for 2nd color. The intensities of both colors are fixed at 3×10^{14} W/cm^2. The carrier envelop phase (CEP) of each of two color fields has been set to be zero.

Figure 1 presents how the ratio of the two-color field changes with respect to the wavelength of the second color for a fixed pulse duration. Note that there are many maxima in the ratio R. Since the ratio is approximately inversely proportional to the duration of isolated attosecond pulses, the two-color fields corresponding to these local maxima are good candidates for high-order harmonic generation with a long continuum. Moreover, we note that the change of ratio with respect to the wavelength is rather dramatic: the ratio falls rapidly from 2.3 to 1.3 when the wavelength increases from 400 nm to 500 nm (Fig. 1A), which suggests that the duration of the attosecond pulse may sensitively depend on the wavelength of the driving laser. Therefore, if a wavelength is properly chosen, ideal isolated attosecond pulses can be obtained.

3. Generation of isolated attosecond pulses

Now that the optimum two-color fields are obtained, the corresponding continuum energy and attosecond pulse duration can be calculated, applying the Lewenstein model [17] to He atoms.

In Fig. 1, the highest ratio of the two-color fields is 2.8 at 1940 nm (the acme of the red line) for 5 fs FWHM. However, this is practically not possible because the period of the 1940 nm color is longer than 5 fs. Thus, this case has to be abandoned.

The second best case is at the ratio of 2.3 (the acme of the blue line) at 1740 nm for 10 fs FWHM, which is practically possible. This two-color field and the corresponding time-frequency diagram produced in the interaction with He are presented in Fig. 2(a) and (b).
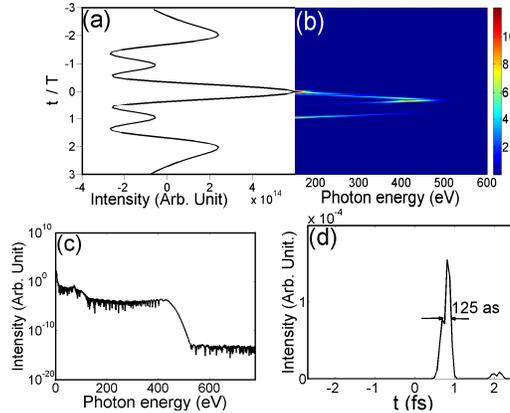

Fig. 2. (a) two-color field of an 800nm, 5fs FWHM, $3 \times 10^{14} W/cm^2$ color mixed with a 1740nm, 10fs FWHM, $3 \times 10^{14} W/cm^2$, T = 2.7 fs (b) time-frequency diagram (c) time-integrated spectrum (d) isolated attosecond pulse produced by a continuum from 440eV to 490eV. The simulation was done for a two-color field interacting with He atom. The carrier envelop phase (CEP) of 800nm and 1740 nm fields has been set to be zero.

As this strong electrical field changes rapidly around $t = 0$, a bound electron experiences a quick ionization and a high acceleration. Thus, the photons at about $t = 1$ fs emitted by this electron possess high energy. The time-integrated continuum is presented in Fig. 2(c), which clearly shows the energy of the continuum to be around 500 eV. What is more, if one is able to select this part of continuum, namely, 440eV~490eV using a multilayer mirror or a filter, an isolated single 125 attosecond pulse is expected to be produced. With a careful chirp management, this pulse can be further shortened.

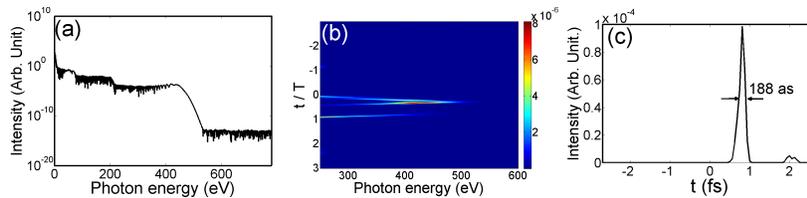There are other two-color fields that are also promising for HHG as Fig. 1 shows.

Fig. 3. Simulation result for the interaction of the two color field of an 800 nm, 10 fs FWHM, $3 \times 10^{14} W/cm^2$ color and a 1820 nm, 15 fs FWHM, $3 \times 10^{14} W/cm^2$ with He. The carrier envelop phase (CEP) of 800nm and 1820 nm fields has been set to be zero. T = 2.7 fs. (a) integrated spectrum (b) time-frequency diagram and (c) isolated attosecond pulse expected from a continuum. An isolated pulse of 188 as is produced by a spectral filter for 470eV~550eV. Helium is used as a working gas medium.

Figures 3(a)-(c) are the results from the interaction of the two-color field of an 800 nm, 10 fs FWHM color and a 1820 nm, 15 fs FWHM with He. And the CEP difference is put at 0. The generated continuums are around 500 eV and an isolated attosecond pulses of about 188 as is expected, when a proper spectral filter for 470eV~550eV is used. Extra chirp management may further shorten them. This case is of great practical meaning since the 15 fs FWHM laser field is currently available by OPA technique.

More simulations were done for local maxima manifested in Fig. 1. The results are listed one by one in Table 1.

Table 1. Various Combinations of Two-Color Fields at the Maxima of R Shown in Fig. 1 with their Resulting Attosecond Pulse Widths and the Energy Range of the Generated Continuum

λ_1 / nm	λ_2 / nm	τ_1 / fs	τ_2 / fs	Cont. energy(eV)	Pulse width(as)
800	350	5	10	100~200	250
800	420	5	5	140~220	313
800	420	5	10	130~220	313
800	420	10	10	140~230	250
800	460	5	10	140~230	312
800	1420	10	15	400~480	313
800	1530	10	10	430~500	375
800	1740	5	10	460~560	125
800	1790	10	10	440~530	125
800	1820	10	15	450~530	188

The laser intensity is 3×10^{14} W / cm^2 and the working gas is He.

As seen in Table 1, if an 800 nm color is mixed with another one from 1500 nm to 1850 nm, high continuum energy can be obtained. However, if an 800 nm color is mixed with a color whose wavelength is below 1000 nm, the continuum energy falls in the range of 100 eV~250 eV. Up to now, a < 5 fs FWHM, 800 nm laser has been used for an isolated attosecond pulse; however, the generation and maintenance of a < 5 fs laser pulse demands extra techniques with care. In this point of view, it is quite encouraging to note in Table 1 that an isolated attosecond pulse can be produced, using 10 fs FWHM, 800 nm light.

4. Comparison between the results from the two-color fields: 800nm + 400nm and 800nm + 1800nm

Since one of the goals of our work is the possibility of using a wider range of wavelength for the second color(300nm-2000nm), we compare the results from the two-color fields 800nm + 400nm and 800 + 1800nm in more details. As the driving field is obtained by a heterodyne mixing of few-optical cycle pulses, the carrier envelop phase (CEP) of each of the two fields should be stabilized. In addition to this, the CEP difference (Δ_{CEP}) between the two field is expected to make an effect. Thus, the influence of CEP difference has been explored. Here we consider four situations: $\Delta_{CEP} = 0$, $\pi/2$, π, $3\pi/2$. The CEP of 800 nm light is varied while the CEP of other colors is fixed at CEP = 0. The results are presented in Table 2. Note that in this simulation, the FWHMs for 400nm, 800nm and 1800nm color are 10fs, 5fs, and 10fs respectively. The intensity for all these three colors are the same as 3×10^{14} W / cm^2 .

Table 2. CEP Effect

Δ_{CEP}	800nm (5 fs) + 400nm (10 fs)			800nm (5fs) + 1800nm (10fs)		
	Cont. energy(eV)	No. of Pulses	Pulse width(as)	Cont. energy(eV)	No. of Pulses	Pulse width(as)
0	125~220	1	313	440~530	1	125
$\pi/2$	125~210	1	250	330~410	1	313
π	100~190	3	-	280~360	1	313
$3\pi/2$	125~205	1	250	335~410	3	-

Continuum energy and attosecond pulses obtained from two color fields, 800nm + 400nm and 800nm + 1800nm in different Δ_{CEP} values. The CEP of the 800 nm light varies as Δ_{CEP} while the CEP of other fields is fixed at 0.

We can observe several points in Table 2. Regardless of Δ_{CEP} change, the continuum energy obtained from the driving field 800nm + 1800nm is always higher than that obtained from the driving field 800nm + 400nm.

It should not be ignored that the continuum energy varies according to Δ_{CEP}. The variation is larger in the case of the 800nm + 1800nm field than in the case of the 800nm + 400nm field. For example, when $\Delta_{CEP} = \pi$, the continuum energy obtained from the 800nm + 1800nm field is in the range of 280eV~360eV; while $\Delta_{CEP} = 0$, it is around 440eV~530eV. The difference is as large as about 160eV. Note also that there may be more than one pulses generated when Δ_{CEP} is not zero. The time-frequency diagrams for 800nm + 1800nm are presented in Fig. 4 which can help us to understand the transition from single to multiple attosecond pulses. From Fig. 4 it can be noted that the stabilization of Δ_{CEP} is important in generating attosecond pulse by two-color mixing. The pulse duration also varies according to Δ_{CEP}. The variation in the pulse duration is also larger for the 800nm + 1800nm field as in the continuum energy. This can be understood in this way. The 10 fs FWHM of 400 light contains ~7.5 cycles; on the other hand, the 10 fs FWHM of 1800 nm light contains 1.8 cycles. The change of the electric field of a laser is more dramatic in 10 fs FWHM, 1800 nm light.
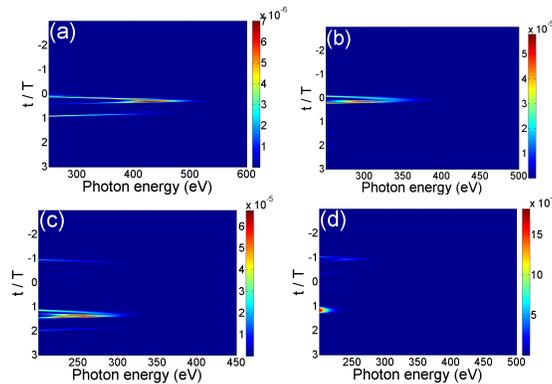

Fig. 4. Time-frequency diagrams for 800nm + 1800nm in different CEP values. Δ_{CEP} for (a)~(d) are $0, \pi/2, \pi, 3\pi/2$ respectively. T = 2.7fs.

5. Conclusion

The generation of isolated attosecond pulses demands the manipulation of the electric field of a laser. DOG and ionization gating are two examples of various manipulation schemes. Here we studied the manipulation by two color mixing. This study covered the wide range of laser wavelengths. The focus was made on the case of equal intensity of two color field. Our study provides a scheme to produce an ~190 attosecond pulse in the spectral region of ~500 eV using 10 - 15 fs FWHM laser fields and identities the laser parameters. These lasers are currently available by OPA technique. The efficiency of the two color mixing process is one of important issues to be addressed. The proper treatment should include the propagation effect and phase-matching issue. This issue is currently under active research for real experiments.