Isolated sub-100-as pulse generation by optimizing two-color laser fields using simulated annealing algorithm

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Abstract: We propose a method to broaden the cut-off region of high harmonic generation by optimizing the parameters of two-color laser fields synthesized by an intense 5 fs pulse at 800 nm and a relatively weak, subharmonic pulse at 2400 nm. Simulated Annealing (SA) algorithm is employed to optimize the electric field amplitude, pulse duration of the control pulse, and the time delay between two pulses. Our simulation shows that a broadened XUV supercontinuum with a 115 eV spectral width can be generated, which is two times broader compared with no optimization, and directly creates an isolated 76as pulse even without any phase compensation.

OCIS codes: (190.4160) Multiharmonic generation; (320.7110) Ultrafast nonlinear optics; (340.7480) X-rays, soft x-rays, extreme ultraviolet (EUV).

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1. Introduction

The advent of attosecond extreme ultraviolet pulses opens up a new regime of tracking ultrafast electron processes in atoms and molecules with unprecedented time resolution [1, 2]. Thus the generation of attosecond pulses has attracted spectacular interest in recent years. A train of attosecond pulses has been observed from HHG process [3], whereas an isolated attosecond pulse is more preferable in XUV-pump-XUV-probe metrology. Although high harmonic generation from a few-cycle driving pulse is a direct method to generate single attosecond pulse [4], sub-100as as pulse is difficult to be generated due to the limitation of current shortest pulse duration. An alternative approach is the temporal confinement of HHG by polarization-gating technique [5,6], which demonstrated to offer 130-as XUV pulses[5], but the single as pulse contains notably less photons than the as pulse trains due to the fact that only a small fraction of the driving pulse energy is used in the generation of as pulses. Very recently, it has been theoretically demonstrated that the attosecond pulse bandwidth can be significantly broadened in a two-color laser field [7,8], because the effect of two-color field is essentially equivalent to shortening the few-cycle pulse duration and maximally utilized the driving laser energy. Z. N. Zeng et al. simulated the HHG using a weak second harmonic control pulse adding upon an intense few-cycle fundamental pulse and generated a supercontinuum with spectral width of 148eV[9]. Meanwhile, Y. H. Zheng et al. experimentally revealed the enhancement and spectral broadening of XUV supercontinuum with suitable phase control [10]. H. Xiong et al. experimentally demonstrated the extension and enhancement of XUV supercontinuum in cutoff region by using the superposition of a weak short pulse and a strong long pulse[11]. Control pulse with other different wavelength such as UV and subharmonic pulse adding upon an intense 800nm pulse have also been intensively investigated [12-15]. However, previous simulations only focus attention on individual physical aspect while ignore optimizing the parameters of two-color fields simultaneously to fully utilize the available laser conditions to obtain spectrally broader and smoother XUV supercontinuum. In this paper, we optimized the electric field amplitude and pulse duration of control pulse, and the initial time delay between the two pulses simultaneously for the first time to generate an isolated sub-100-as pulse.

2. Theoretical details and simulation results

HHG process from the interaction of noble gas with a strong laser pulse is well understood by the three-step model: the bound electron first tunnels through the barrier formed by the Coulomb potential and the laser field, then it travels almost freely in the oscillating laser field, finally, it recombines with the parent ion in some probability, emitting photons. The emitted photon energy is equal to the ionization potential plus the kinetic energy that the electron gained in the laser field, and the kinetic energy can be classically calculated as $E_{ki} = \int_{t_i}^{t_r} E(t)dt$, where t_i and t_r are the electron ionization and recombination time. The maximum kinetic energy of an electron returning to its parent ion is different for each half optical cycle in the few-cycle laser field since the electric field amplitude varies significantly from one half cycle to the next. Thus the bandwidth of the XUV supercontinuum is proportional to the gap between the highest and the second highest half-cycle cutoff photon energies in the few-cycle laser field. We choose the energy difference between the highest

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E_{kimax} and the second highest $E_{kimax'}$ as the target function $G=E_{kimax}-E_{kimax'}$ in our SA method.

The adopted simulated annealing method is a Monte Carlo approach for minimizing multivariate functions, based on the principles of thermodynamics. This algorithm has been theoretically testified to have gradual convergence, and under certain conditions, it can find the global minimum, approaching the best solution of the entire solution domains. The procedure of the SA algorithm is described in detail as follows:

(1) Define some initial parameters used in the SA algorithm, such as the high initial temperature T_0, dropping rate of temperature η, and target function G. Start from a random solution S, where S is a series of numerical digits of laser parameters.

(2) Randomly generate new solution S', calculate the associated function cost △G'=G(S')-G(S), where G(S) is the target function.

(3) If △G'<0, i.e., the cost is lower, then this new solution is accepted. Otherwise, the new solution is accepted or rejected with a probability of exp(-△G'/kT), where k and T are the Boltzmann constant and current temperature. If S' was accepted, then S(q+1)=S, otherwise, S(q+1)=S(q). This procedure is the famous metropolis process.

(4) Decrease the temperature as $T=\eta*T_0$ and continue procedure 2 and procedure 3 until the current solution is the best and end the calculation. Generally speaking, the qualification for ending the calculation is that tens of new solutions are rejected consecutively.

The model of HHG simulation is based on single-active atom approximation [4]. The atomic dipole moment is calculated in Eq. (34) in reference [4], and the high harmonic spectrum is obtained by Fourier transforming the time-dependent dipole moment. In the simulation, we take argon (Ar) as the model atom, and choose a Ti:sapphire laser pulse with a wavelength of 800nm, pulse duration of 5fs and an intensity of 3×10^{14}W/cm^2, and a weak control pulse with wavelength of 2400nm. Technically, a 2400nm laser pulse can be generated with an optical parametric amplifier (OPA). The fundamental laser pulse and subharmonic pulse are both linearly polarized and the synthesized electric field can be given by

$$E_s = E_1 \exp[-2\ln(2)t^2/\tau_1^2]\cos(\omega t) + E_2 \exp[-2\ln(2)(t-t_0)^2/\tau_2^2]\cos[\omega/3(t-t_0)] \qquad (1)$$

here E_1 and E_2 are the amplitudes of the electric fields of the fundamental and the subharmonic control pulses, respectively; ω is the frequency of the fundamental pulse; and τ_1 and τ_2 are the corresponding pulse durations (FWHM). The t_0 in Eq.(1) defines the initial time delay between the fundamental and the subharmonic pulses, creating a relative phase delay of $2\pi t_0/3T$, where T is the optical period of the fundamental wave.

To make comparison and illustrate the advantage of optimization method, we carried out our simulation in the following three different cases: firstly, using only 5fs driving pulse; secondly, superimposing a 64fs, 2400nm pulse with an intensity of 3×10^{13}W/cm^2 onto the driving pulse, and thirdly, optimizing the parameters of the two-color laser pulses. Fig. 1. shows the HHG spectra generated in the three optical fields. The XUV supercontinuum spectrum in the cutoff region produced by the single 5fs pulse alone shows a bandwidth of approximate 10eV, as shown by the black curve in Fig. 1. A significantly broadened XUV supercontinuum with a spectral width of ~60eV is achieved by adding the weak 2400nm pulse onto the fundamental pulse with zero time delay, as can be seen in Fig. 1 (blue curve). In this case, the contribution to the broadening of the XUV supercontinuum spectrum is mainly from the extension of the cutoff region. Recent theory point out that significant extension of the cutoff high harmonic energy in two color field is a result of the heterodyne mixing of laser fields, and our results is well consistent to this physical picture[7]. For generating attosecond pulses with sub-100-as duration, XUV supercontinuum with broader spectral widths are required. By using the SA algorithm, we optimized three parameters of the two pulses

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simultaneously in the available scope of laser conditions, and got a dramatically increased XUV supercontinuum with spectral width of 115eV, which is nearly two times broader compared with no optimization. The optimized control pulse have an intensity of 7.4×10^{13}W/cm^2, pulse duration of 44.8fs, with time delay of -4fs respect to the fundamental pulse, and these laser parameters can be easily facilitated in current laboratories. Thus we can see that the quantum path control of the HHG process must be more accurately manipulated and the more broadened XUV supercontinuum spectrum can be generated by using optimizing method.

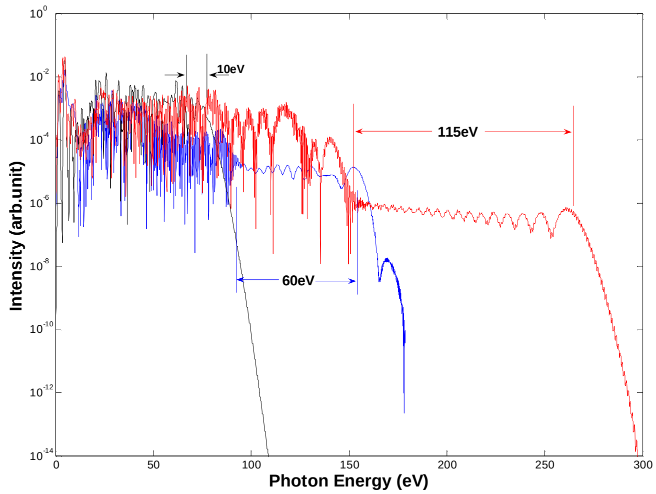

Fig. 1. The spectra of XUV supercontinua generated with the single 5 fs pulse (black), the initial two-color pulses (blue), and with the optimized two-color field (red).

Now we examine the temporal profile of the as pulse supported by the broadened XUV supercontinuum in Fig. 1. (red curve). A simple inverse Fourier transformation of the XUV supercontinuum in the spectral range of 150-265eV gives out an isolated 76as pulse even without any dispersion compensation, as is presented in Fig. 2. And it can be theoretically predicted that if the attochirp over the 115eV spectral range can be properly compensated for by appropriate metal filters, thus the isolated as pulse duration could be more short and have a more clean temporal profile.

We also performed the time-frequency analyses of the dipole responses of the argon atom to the three different optical fields [16], as shown in Fig. 3, to obtain more physical information behind the dramatic extension of the cutoff region in the optimized two color laser field. The X-axis and Y-axis of the time-frequency diagram show the energies and emission times of photons, respectively. In order to clearly see the difference of harmonic spectrum from the three different optical fields, we pick three peaks named as P1, P2, P3 in Fig. 3 (a), (b), (c), respectively. The three peaks present the maximum kinetic energies of the electrons returning to their parent ions. It can be seen from Fig. 3(a) that there are two arms of comparable intensities of the XUV radiation in P1, corresponding to the long and short trajectories in the HHG process. The long and short trajectories generally interfere with each other, which could result in multi-peak structures in the generated as XUV pulses. With the use of 2400nm control pulse adding to the fundamental pulses, only one quantum path is picked out. As shown in Fig. 3(b), P2 is stronger than other peaks, and the cutoff energy difference between the two adjacent peaks is 60eV. After optimizing the parameters of these two-color laser pulses, XUV supercontinuum with spectral width of 115eV is obtained, as presented in Fig. 3(c). In this optimized case, P3 is picked out while other peaks are greatly suppressed, thus dramatically broadened XUV supercontinuum spectrum can be generated.

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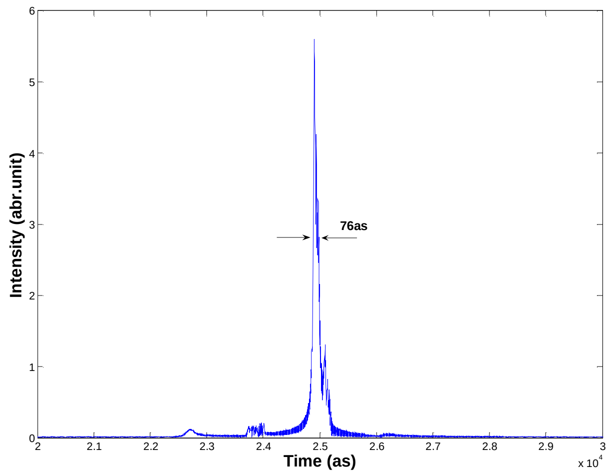

Fig. 2. The temporal profile of the single XUV pulses generated in the optimized two-color field without any phase compensation.

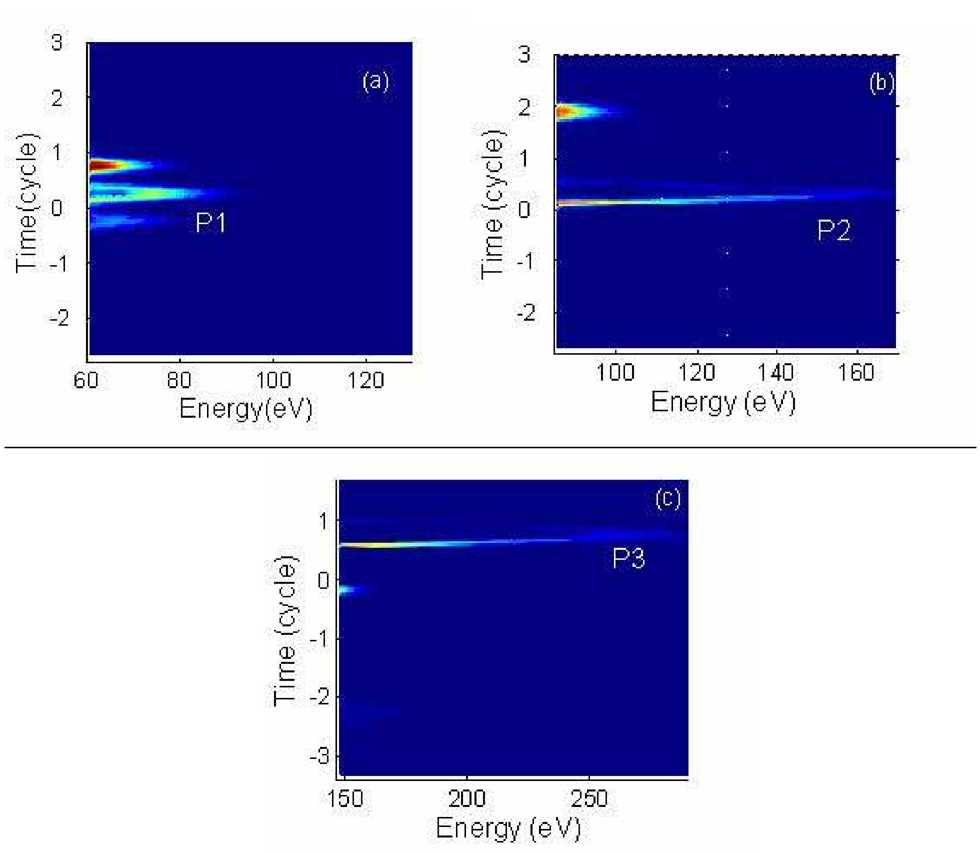

Fig. 3. Time-frequency diagrams of the XUV supercontinuum spectra generated by (a) the single-color 5fs pulse, (b) the two-color pulses, and (c) the parameter optimized two-color pulses.

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3. Conclusion

In conclusion, an optimizing method for efficient generation of an isolated attosecond pulse in a two-color field has been proposed. It is shown that a broad XUV supercontinuum with 115 eV spectral width can be generated by optimizing the electric field amplitude, pulse duration of the control pulse, and the time delay between the two pulses. The optimized spectral width of XUV supercontinuum is nearly two times broader compared with no optimization and directly creates an isolated 76as (FWHM) pulse even without any phase compensation. As can be seen that by using the optimization method, we can maximally utilize the available laser conditions to generate broader and smoother XUV supercontinuum spectrum by optimizing the laser parameters that can be easily manipulated.

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