Controls for the generation of high-order harmonics and attosecond pulses by an infrared laser field combined with a low-frequency pulse*

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(Received 7 December 2011; revised manuscript received 20 January 2012)

We investigate high-order harmonic generations by controlling various quantum paths of harmonics in an infrared laser field which combines a low-frequency pulse. Both classical theory and the quantum wavelet transform method are used to understand the physics of harmonics. By adjusting the carrier envelope phase of the fundamental field, the intensities of harmonic spectra increase and the harmonics in the plateau become regular. Attosecond pulses each with a duration of 58 as are obtained directly by compressing the harmonics, and with phase compensation an isolated attosecond pulse less than 30 as can be generated.

Keywords: high-order harmonics, isolated attosecond pulse, LZH-DICP code

PACS: 02.60.Cb, 32.80.Rj, 42.65.Ky **DOI:** 10.1088/1674-1056/21/8/080202

1. Introduction

Great interest has been aroused in the high-order harmonics generation (HHG) due to their potential for the generation of an extreme ultraviolet (XUV) light source and attosecond pulses, which may enable us to solve electronic dynamics[1] just as subpicosecond or femtosecond pulses permit the observation of molecular motions and Rydberg orbital motions in real time. HHG has been well-explained by Corkum[2] using the semiclassical theory, also named simple-man's model: when atoms or molecules are exposed to an intense laser field, the electron first escapes and accelerates, then decelerates to zero velocity and turns back when the laser field changes its sign, it finally recombines with the parent ionic core, and simultaneously emits photons each with an energy equal to the ionization potential plus the kinetic energy which the electron gains from the laser field. The corresponding harmonic spectrum intensity decreases rapidly at low orders, then exhibits a plateau region and decreases again with a cutoff at a well-known energy of $I_p + 3.17U_p$, where U_p ($U_p = I/4\omega^2$) is the ponderomotive energy. Further, Lewenstein *et al.*[3] confirmed this cutoff law by quantum theories. This process represents an extreme limit of nonlinear optics, where thousands of visible photons each with an energy of 1 eV–2 eV are combined together, resulting in a coherent beam with photon energies up to a few keV. And it is repeated every half cycle of the laser field, during which an attosecond pulse train is emitted when multicycle pulses are employed. However, when the symmetry of the laser field is broken, an isolated attosecond pulse can be obtained directly which has much more applications, thus attracting lots of attention. Recently, Kienberger[4] generated a single attosecond pulse with a duration down to 250 as by using a few-cycle laser pulse with a duration of 5 fs. Lately, an isolated attosecond pulse with a duration of 130 as has been generated via the chirp compensation technique.[5] Furthermore, interaction between matter and a single wave cycle light has been investigated which resulted in an 80-as isolated pulse.[6] So far, two-colour laser fields have been adopted generally to gain an even shorter single attosecond pulse.[7] An isolated attosecond pulse was obtained by adding weak frequency-doubled pulses into strong fundamental pulses at a relative phase of 1.5π.[8] Moreover, a high-order harmonic has also been simulated in a two-colour laser field by a weak fundamental pulse with a strong frequency-doubled laser pulse.[9] A broad flat XUV supercontinuum was produced by a strong control laser pulse plus its second harmonic field whose intensity was also strong, and

*Project supported by the National Natural Science Foundation of China (Grant No. 10974198).

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the duration of the generated attosecond pulse could reach one atomic unit of time (24 as) using a phase compensation method.[10] However its width was at the expense of the intensity of the harmonic. Kim *et al.*[11] increased the transfer efficiency of the high-order harmonic generation using an orthogonally polarized two-colour laser field. Subsequently, a single 350-as pulse has been generated using these laser fields by selecting a pulse with a polarization direction of 167°.[12] Besides, the harmonic generation efficiency could be improved by controlling the phase-matching of HHG in a two-colour laser field.[13] In addition, an extreme ultraviolet supercontinuum can be generated by a three-colour laser field.[14,15] HHG has also been studied by two laser pulses using a pump-probe technique experimentally.[16]

2. Theory and discussion

In this paper, we add a relatively weak low-frequency pulse with the same envelope as the fundamental field. The expression of the synthesized field can be written as

$$E = E_1 \exp[-4\ln(2)t^2/\tau_1^2]\cos(\omega_1 t + \phi) + E_2 \exp[-4\ln(2)t^2/\tau_2^2]\cos(\omega_2(t+t_0)),$$

where E_1 and E_2 are the amplitudes of the electric fields; τ_1 and τ_2 are the pulse durations of the fundamental and the low-frequency pulse, respectively. The intensity of the fundamental field is 5×10^{14} W/cm^2. We scan the intensity of the low-frequency pulse and find that when it is 5×10^{13} W/cm^2, the longest bandwidth of the continuum could be obtained. The Gaussian profile is adopted. ϕ is the carrier-envelope phase (CEP) of the fundamental field. We choose $\omega_1 = 0.057$ and $\omega_2 = 0.003$, corresponding to the wavelengths of 800 nm and 15 μm, respectively, and ignore the time delay between these two laser fields, which means $t_0 = 0$. Figure 1 shows the three electric fields of the single 5-fs fundamental pulse, the 20-fs low-frequency pulse, and the synthesized field. The positive amplitude of the synthesized field increases in this time region because the low-frequency field is always positive during the time of the fundamental field due to the fact that the paths of electrons moving in the laser field have been changed. The kinetic energies which electrons gain from the laser field and are dependent on ionization and recombination time, are shown in Fig. 2. It should be noted that the wave packets which first ionize around peak A will then gain energy from the following peak field B and return to the parent ionic core between B and C (approximately 2/3 optical cycle) as shown in Fig. 2(a). In the single laser field, the tunneling electron wave packets launch around each peak and recollide with the ionic core, which process repeats every half cycle. However, it can be seen that only the electrons ionized near peak B and peak D in Fig. 1 can recollide with the ionic core in the synthesized field. The additional low-frequency laser pulses significantly modulate the maximum kinetic energy so that the cutoff energy extends up to $8U_p$. Simultaneously, only the short trajectory is reserved, which is convenient for controlling the relative phases between the harmonics, up to the production of the single attosecond pulse. The bandwidth of the continuum produced by the single fundamental field shows as approximately 30 eV as indicated in Fig. 2(a), which is significantly broadened to more than 190 eV by

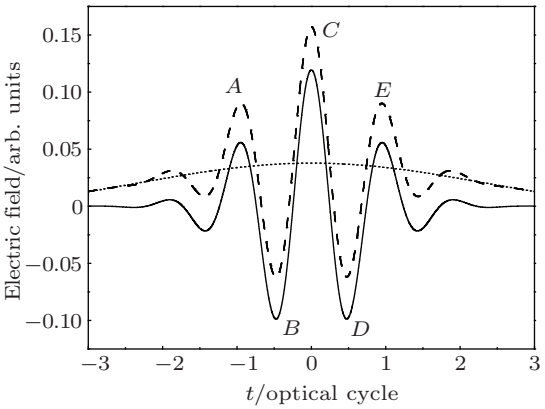


Fig. 1. Electric fields for single 5 fs/800 nm (solid line), low-frequency 20 fs/15 μm (dotted line), and the synthesized pulses (dashed line).

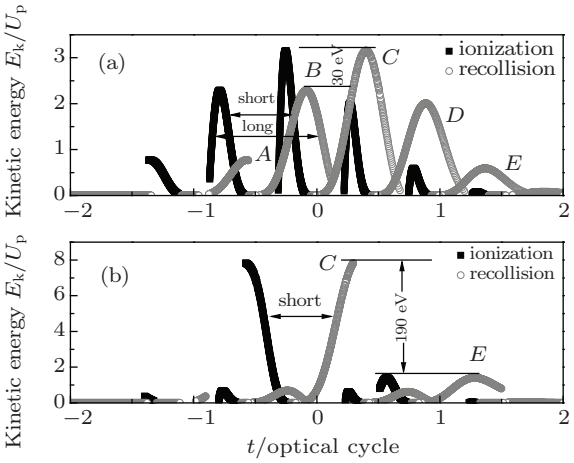


Fig. 2. (a) Electronic kinetic energies dependent on ionization (black square) and recollision time (gray circle) for single 5-fs pulses by the classical theory. (b) The same as (a) but for the synthesized pulses with $\phi_{\rm CEP} = 0$.

the synthesized field shown in Fig. 2(b). Thus the minimum duration of the generated attosecond pulse could be less than 30 as in the Fourier transform limit that it is much powerful for solving electron dynamics.

We consider a helium atom system and simulate the harmonic spectra by numerically solving the time-dependent Schrödinger equation through using a split-operator approach for the propagation of the wavepacket.[17−19] The LZH-DICP code[15,20−23] is used in our simulations. The Coulomb potential is represented by the 'soft-core' potential $V(x) = -1/\sqrt{\varepsilon + x^2}$ and we choose the soften parameter $\varepsilon = 0.484$ corresponding to an ionization energy of 24.6 eV for the ground state. The harmonic spectra are obtained by Fourier transform of the time-dependent dipole acceleration. Figure 3 shows the three harmonic spectra by the single 5-fs laser field, and synthesized field with carrier envelope phases $\phi_{\rm CEP} = 0$ and 1.4π, respectively. For $\phi_{\rm CEP} = 0$, the quantum results are quite in agreement with classical results that the cutoff region is broadened to more than 190 eV by the synthesized field, and an isolated attosecond pulse whose duration is less than 30 as can be obtained if harmonics are phase-matched. However, it is also shown that the harmonic intensities of the cutoff region are almost two orders of magnitude lower than that for the single pulse, leading to low transfer efficiency of the production of attosecond pulses. Otherwise, the harmonics in the cutoff regime are irregular in the continuum region. Actually, the CEP effect should be considered since the duration of the control laser field is very short. We test the CEP effect, which shows that the results for $\phi_{\rm CEP} = 1.4\pi$ are much better than others. In Fig. 3, the continuum in the spectrum is very regular for $\phi_{\rm CEP} = 1.4\pi$ compared with that for $\phi_{\rm CEP} = 0$, and the intensities of the harmonic spectra all increase significantly.

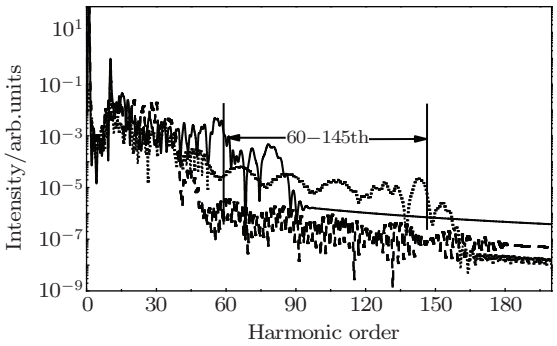


Fig. 3. High-order harmonic spectra obtained from the single pulse (solid line) and the synthesized pulse with $\phi_{\rm CEP} = 0$ (dashed line) and $\phi_{\rm CEP} = 1.4\pi$ (dotted line).

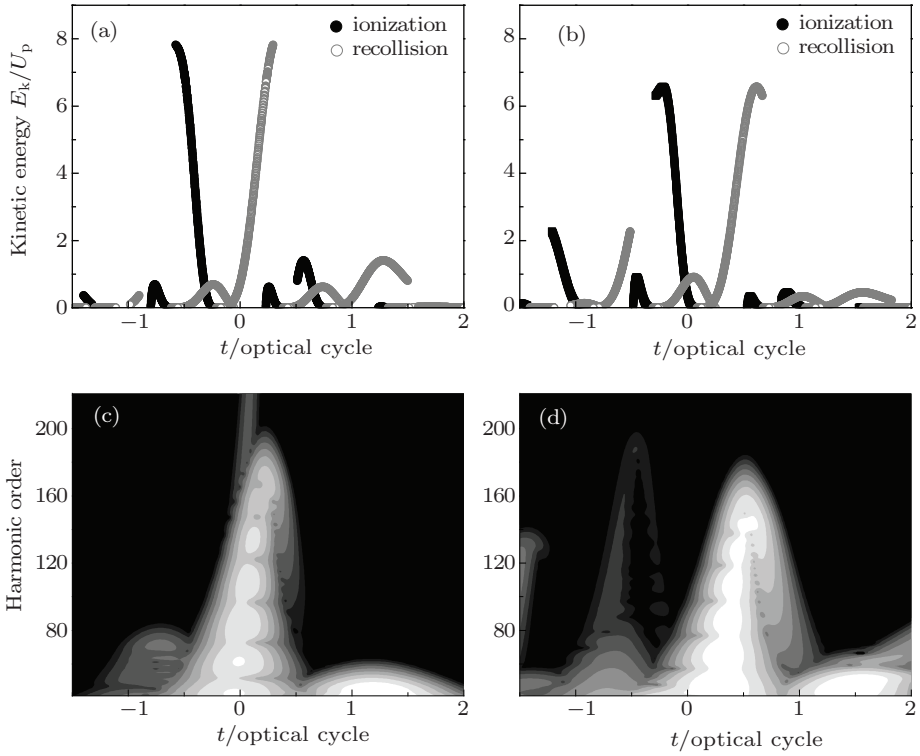


Fig. 4. Electronic kinetic energy dependent on ionization (black square) and recollision (gray circle) time for the synthesized pulse with (a) $\phi_{\rm CEP} = 0$ and (b) $\phi_{\rm CEP} = 1.4\pi$. Corresponding time-frequency analyses of harmonics are shown in panels (c) and (d), respectively.

Classical trajectories and time-frequency analyses of the dipole responses[24−26] with different CEPs are presented in Figs. 4. It is shown that the quantum results are quite consistent with those from the classical simulations by simply solving the Newton's equation. Obviously, the plateau region from about the 45th order to the 170th order for $\phi_{\rm CEP} = 0$ is broader than that for $\phi_{\rm CEP} = 1.4\pi$ which is from about the 60th order to the 145th order. A similar pattern can be observed in that only one main channel (short trajectory) exists in the region for both by classical theory. For accuracy, a quantum wavelet transform method is adopted, thereby not only indicating the emission channels, but also providing the relative intensities between the channels. Figure 4(d) shows the intensities of the spectra in the continuum region for $\phi_{\rm CEP} = 1.4\pi$ which are much more powerful than those for $\phi_{\rm CEP} = 0$ in Fig. 4(c).

Figure 5 indicates the temporal profile of an attosecond pulse generated by combining 60th-to-145th order harmonics in the synthesized field with $\phi_{\rm CEP} = 1.4\pi$. An attosecond pulse with a duration of 58 as is obtained directly, close to the Fourier transform limit without phase compensation. There are many lower peaks besides the main one, owing to phase mismatching of the harmonics. However, many ways are available to modulate the phases between harmonics, such as, chirp compensation,[6] grating-assisted phase matching,[26] etc. The solid line in Fig. 5 shows the result with phase compensation that the duration reaches 26 as nearly to the one atomic unit of time, which is powerful for the investigation of ultrafast dynamics. This short pulse duration indicates that adding a low-frequency field is an efficient method of producing a single attosecond pulse from HHG in noble gases.

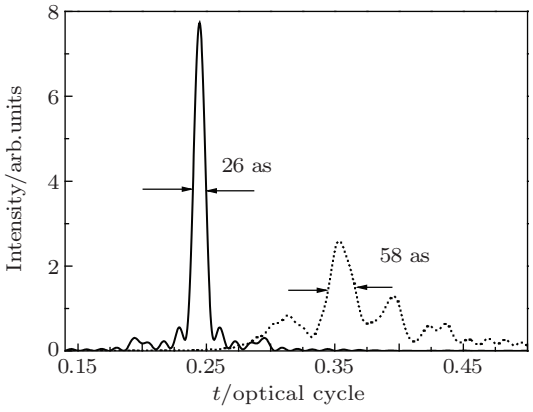


Fig. 5. Temporal profiles of an attosecond pulse generated from 60th to 145th order harmonics (shown in Fig. 3) with (dotted line) and without (solid line) phase compensation.

3. Conclusion

In this paper, we demonstrate that the synthesized laser field whose fundamental field is combined with a low-frequency laser field leads to a significant modulation of the high order harmonics and attosecond pulse generation. Only the short trajectories of the quantum paths exist in the plateau region. By adjusting the CEP, not only the intensity of harmonic is increased, but also more regular harmonics are obtained in the continuum region which has advantages for the generation of the attosecond pulse. With the phase compensation technique, a single attosecond pulse with a duration of 26 as that is nearly at the Fourier transform limit is obtained, which has much more applications in probing electronic dynamics.

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