Quantum Control for Broadband Attosecond Pulse Generation

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In our work, we propose a scheme for coherent control the HHG with an ultrashort laser pulse in combination with a controlling pulse. It is shown that this method can significantly modulate the electron dynamics and a broadband supercontinuous high harmonics can be generated. Then an isolated sub-100 attosecond pulse can be obtained, which allows one to measure a wide range of ultrafast dynamics not normally accessible before.

Key Words: High harmonic generation, Isolated attosecond pulse, Two-color field

1. Introduction

Driven by intense laser pulses, atomic and molecular systems can emit lights at frequencies multiple of that of the laser field. This nonlinear process, known as high harmonic generation (HHG), is a subject of great interest owing to its potential applications for the coherent extreme ultraviolet source and the generation of attosecond pulses. Attosecond extreme ultraviolet pulses, especially the isolated attosecond pulse, allow one to investigate ultrafast electronic processes, opening the door to attophysics. The straightforward attosecond metrology prefers an isolated attosecond pulse rather than a train of attosecond pulses.¹⁻²⁾ Hence much effort is paid to produce the isolated pulse.¹⁻⁵⁾ If using a few-cycle pulse, the highest order harmonics emits only at the peak of the laser pulse and cutoff region of the spectrum becomes a continuum, then an isolated attosecond pulse can be filtered out. This scheme has been experimentally carried out by using a state-of-art few-cycle laser pulse. 1-3) However, the bandwidth of the harmonics in the cutoff region is only about 20 eV, therefore the duration of shortest attosecond pulse is about 250 as, ¹⁾ which is greater than the natural timescale of the electronic process inside atoms (152 as, i.e., the period of electrons in the Bohr orbit of ground-state hydrogens), and then the application of the 250-as pulse is limited. It has been reported that broadband continuous harmonics can be observed using a few-cycle driving pulse with the polarization gating technique. ⁶⁾ Recently, this scheme has been experimentally achieved by Sansone et al. ⁷⁾ In their experiment, a single cycle attosecond pulse with the duration of 130 as is generated with the chirp compensation technique. High harmonic generation (HHG) is well understood in terms of the semiclassical three-step model ⁸, i.e., the electron is first ionized, then is accelerated in the laser field, finally the electron recombines with the parent ion. One can control the HHG by manipulating different steps. In this work, we propose a new facile method for isolated sub-100-attosecond pulse generation via controlling electron dynamics (the second step) using a few-cycle laser pulse in combination with a controlling field.

2. Results and discussions

We first investigate the HHG process with the semiclassical model, ⁸⁾ which gives us a clear physics picture. The electric

field is given by $E(t) = f(t)[E_0\cos(\omega_0 t + \varphi_0) + E_1\cos(\omega_1 t)]$. E_0 and E_1 are the amplitudes, ω_0 and ω_1 are the frequencies of the driving and controlling fields, respectively, φ_0 is the carrier envelope phase, f(t) is the pulse envelope. Figure 1(a) illustrates the sketch of HHG process from atoms driven by a few-cycle laser pulse with $\varphi_0 = 0$. We choose helium as the target gas, whose ionization energy is 24.5 eV. As shown in the figure, the driving pulse contains only two optical cycle, thus the electron is mainly ionized near the peaks of the electric field and forms three dominant returns(marked as R₁,



Fig. 1 (a) A sketch of electron dynamics in a 5-fs laser pulse. (b) The dependence of kinetic energy of electron on the ionization (dots) and recombination times (cross). The laser intensity and wavelength are $8.3 \times 10^{14} \text{ W/cm}^2$ and 800 nm.

 R_2 and R_3). Figure 1(b) presents the dependence of the kinetic the ionization(dots) and recombination energy on times(crosses). It is shown that there are two classes of trajectories corresponding to the same energies of the returning electrons in each half optical cycle. The first trajectory with earlier ionization but later recombination times is called long trajectory, and the other one with later ionization but earlier emission times is called short trajectory. The maximum kinetic energies of R₁, R₂ and R₃ are 140, 160 and 130 eV, respectively. Therefore only R2 contributes to the harmonics higher than 140 eV, which can be filtered out to generate an isolated attosecond pulses. However the bandwidth of this pulse is only 20 eV, and the minimum pulse duration is about 250 as

By adding a controlling field (with a different frequency) to the few-cycle driving field, the electron dynamics can be modulated. The dotted line in Fig. 2(a) shows the 5-fs driving laser pulse, the dashed line shows the controlling field with a frequency of $\omega_1 = 2\omega_0$, and the solid line is the synthesized field. The intensity of the driving field is 8.3×10^{14} W/cm² and the controlling field is 4% of the driving field. By adjusting the relative phase, the controlling field can be set in the same direction with the driving field in the half cycle of t = 2.5T0(the return *R*2), then the electron will gain much higher energy since the driving field is enhanced. In the adjacent half cycles (the returns *R*1 and *R*3), the driving and controlling fields change their directions and are in opposite directions, then driving field is weakened and the electron gains less energy.



Fig. 2 (a) The electric fields of the synthesized field (solid line) of a 5-fs driving laser pulse (doted line) in combination with its second-harmonic controlling pulse (dashed line). The intensity of the controlling field is only 4% of the driving field and the relative phase (b) The dependence of kinetic energy of electron on the ionization (dots) and recombination times (cross) in the synthesized field shown in (a) The driving laser intensity and wavelength are 8.3×10^{14} W/cm² and 800 nm, $\varphi_0 = 0.2\pi$.



Fig. 3 Same as Fig. 2, but for a laser field with an intensity of 6 x 10^{14} W/cm² and pulse duration of 10 fs. $\omega_1 = 0.5 \ \omega_0$ and $\varphi_0 = 0$.

This larger contrast between the neighboring half-cycles will broaden the bandwidth of the continuous harmonics in the cutoff, which leads to an isolated broadband attosecond pulse. Figure 2(b) shows the dependence of the kinetic energy on the ionization (dots) and recombination times (cross) in the synthesized field. In contrast to Fig. 1(b), one can see from Fig. 2(b) that the bandwidth of the high harmonics in the cutoff is broadened up to 60 eV, corresponding to an isolated attosecond pulse of about 70 as in the Fourier transform limit.

There are many degrees of freedom for choosing the controlling field. Figure 3(a) and (b) show the HHG in the 10-fs driving laser pulse in combination with a sub-harmonic controlling field ($\omega_1 = 0.5 \omega_0$). The intensity of the driving laser pulse is 6 x 10¹⁴ W/cm², and the controlling field is 20% of the driving field. One can see that an isolated attosecond pulse with a bandwidth of 150 eV will be generated, which corresponds to an isolated 25-as pulse in the Fourier transform limit. Figure 4(a) and (b) show the HHG in the 5-fs driving laser pulse in combination with a low-frequency field ($\omega_1 = 0.08 \omega_0$). One can see that a broadband high harmonics of about 140 eV can be obtained. It is worth noting that the results in Fig. 3 are obtained using a 10-fs laser pulse. In contrast to the 5-fs pulse shown in Figs. 2 and 4, the 10-fs laser system is relatively easier achieved.

Following, we investigate the harmonics and the attosecond pulse generation by numerically solving the time-dependent Schrodinger equation by means of the split-operator method.⁹⁾ Figure 5(a) shows the harmonic spectrum in the combination of the 10-fs driving pulse and the sub-harmonic controlling field. One can clearly see that the spectrum shows the structure as expect in our semiclassical approach: it is irregular for the low harmonics and fascinatingly becomes regular and continuous from 160th harmonic to 230th harmonic. It is because many returns contribute to the low harmonics and the interference of these returns leads to a chaotic structure. For



Fig. 4 Same as Fig. 2, but for a laser field with an intensity of 8 x 10^{14} W/cm² $\omega_1 = 0.08 \omega_0$ and $\varphi_0 = 0$.

the highest harmonics, only one return (*R*2) contributes to the HHG, hence a regular and continuous structure is present. As shown in Fig. 5 (a), the bandwidth of the continuous harmonics is significantly broadened up to 105 eV and an isolated attosecond pulse with a duration of 70 as is produced (see Fig. 5 (b)). Figure 6 (a) shows the harmonic spectrum in a 5-fs pulse by mixing a low-frequency field of 0.08 $a_{\rm b}$. One



Fig. 5 (a) High harmonic generation in an ultrashort driving pulse by mixing a sub-harmonic controlling field. (b) Attosecond pulses generated by selecting the supercontinuous harmonics in the cutofff. The parameters are the same as in Fig. 3.



Fig. 6 (a) High harmonic generation in an ultrashort driving pulse by mixing a low-frequency controlling field. (b) Attosecond pulses generated by selecting the supercontinuous harmonics in the cutofff. The other parameters are the same as in Fig. 4.

can clearly see that the harmonic spectra show the similar structure as shown in Fig. 5(a) and a supercontinous high harmonics with a bandwidth of about 155 eV are generated. By selecting these high harmonics, an isolated 48 as pulse is produced.

3. Summary

In summary, we propose a new method for coherently controlling the electron dynamics using a few-cycle laser pulse in combination with a controlling field. It is shown that this method can broaden the bandwidth and then an isolated sub-100 attosecond pulse is straightforward obtained. Such an ultrashort pulse allows one to investigate ultrafast electronic processes which have never been achieved before and to manipulate the electronic dynamics upon changing the pulse phase in the extreme ultraviolet regime. In addition, the few-cycle synthesized pulse allows one to manipulate the electron dynamics more powerfully, which may be used to control the other laser-atom interaction processes.

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