High-order harmonics generation by few-cycle and multi-cycle femtosecond laser pulses*

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This paper investigates experimentally high-order harmonic generation (HHG) of neon gas with 5-fs and 25-fs driving laser pulses. It has been demonstrated that the cutoff energy of the harmonic extreme ultraviolet photons is extended to 131 eV and the HHG spectrum near the cutoff region becomes continuum as the driving laser pulse duration is 5 fs; whereas much lower cutoff photon energy and discrete harmonic spectrum near the cutoff region are presented as the laser pulse duration is 25 fs. The results can be explained by the fact that neutral atoms can be exposed to more intense laser field before they are depleted by ionization because of the extremely short rising time of the few-cycle pulse. The 5-fs driving laser pulse paves the way of generation of coherent x-ray in the water window and single attosecond pulse.

Keywords: high harmonics generation, short pulse, super-continuum spectrum

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1. Introduction

With the development of few-cycle laser pulses, the interaction between intense few-cycle laser pulses and atoms, molecules, and solid materials has become a very attractive research area.[1,2] It is well accepted that the high harmonic generation (HHG) can be described by a three-step model: tunneling, acceleration and recombination. The tunneling rate of an atom is governed by the amplitude of the laser's electric field, and the acceleration is determined by the electric field oscillation, which predicts the cutoff energy of the HHG as $\hbar\omega_{\rm cutoff} = I_{\rm p} + 3.2U_{\rm p}$ under monochromatic field, where $I_{\rm p}$ is the ionization potential of the target, and $U_{\rm p}$ is the ponderomotive energy of free electron in the laser field.[3] When the driving laser consists of only few laser cycles, the rising time of the laser field reaching the intensity required for tunnel ionization is very short, while in this case the atom can survive to higher laser intensity prior to ionization. The neutral atoms can be exposed to a stronger and rapidly increasing laser field before they are depleted by ionization, which allows the electron to gain more energy before recollision with parent ion.[4−6] This is called non-adiabatic effect in HHG when the driver laser is few-cycle pulse. When the driver laser is a long pulse, for example 25 fs, which contains many optical cycles with almost the same electric amplitude, this implies that the field amplitude changes little between successive optical cycles. Thus when the laser pulse reaches the intensity required for tunneling ionization, there is sufficient time for the electron to escape from the atomic core before the laser intensity increases further.[7] The HHG emission occurs at lower laser field amplitude and kinetic energy of electron increases slowly from laser field, which results in the generation of weak and lower energy harmonic peaks.[8] Therefore, higher saturation laser intensity will be achieved by using shorter laser pulse, and results in higher ponderomotive energy and more intense harmonics emission than that by using longer pulses.[9] The short laser pulse, especially for few-cycle pulses, has the potential of generation of single attosecond pulse and soft x-ray in "water window" (2∼4 nm).

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The pulse duration down to sub 5 fs with high energy is achieved from hollow fibre and chirped mirror compressor in our laboratory.[10] We carried out the measurement of high harmonic spectra generated from 5-fs pulses and 25-fs pulses. The experimental results show that the highest energy of HHG is extended to ∼131 eV and spectral distribution at cut-off region is smooth and continuous which covers from 22 nm to 9 nm for 5-fs driving laser pulses, whereas discrete harmonic peaks and lower energy extreme ultraviolet (XUV) emission for 25 fs. These results are in agreement with the theoretical studies, which indicate that the dramatically shortened rise time of the laser pulse (< 2 optical cycles) allows atomic dipole radiation to be produced at unprecedentedly low ionization probability, extending coherent growth of XUV harmonics to higher photon energy.

2. Experimental setup

A standard Ti: sapphire chirped pulse amplifier (CPA) produces 800-nm optical pulses with energy of 0.8 mJ in 25 fs at repetition rate of 1 kHz. The pulses are focused into hollow fibre with differential pumped neon gas to get spectral broadening, and subsequently chirped mirror compressor and finely tunable small angle wedge, the shortest pulse duration of less than 5 fs with energy of 0.5 mJ is achieved, the details of laser is described in Ref. [10].

The neon gas target is formed by a thin metallic tube having a wall thickness of 0.05 mm and an inner diameter of 3 mm, which is squeezed to about 1 mm. The laser beam is focused with a silver-coated spherical mirror (focal length, 350 mm) through a 1-mm thick fused silica window into a vacuum chamber containing the target at the focus. The hole in the tube wall is bored by the laser beam itself, minimizing the gas load to the surrounding vacuum. The pulse energy is ∼0.33 mJ on the target, and is focused to a $1/e^2$ diameter of 60 μm, implying a peak intensity of $\sim 10^{15}$ W/cm^2 on the beam axis. The tube is continuously backed with pressures of 200∼250 mbar. The vacuum of target chamber is about $\sim 6.4\times 10^{-2}$ mbar when gas is working. The XUV radiation produced collinearly with the laser beam passes through a 1-mm aperture (placed ≈30 cm behind the target) into a 1-m grazing incidence flat-field spectrometer. The fundamental light is blocked by a metallic foil, which transmit the photon energy band of interest. The foils are originally designed for greater photon energies, and they are related to different wavelength ranges. The flat-field spectrometer, which is equipped with 1200 lines/mm (mean value) varied line spacing concave grating and soft x-ray coupled charge device (CCD) (PIXIS-XO, Princeton), is evaluated to be 10^{-4} mbar.

3. Results and discussion

In preliminary experimental studies, in order to demonstrate the resolution and calibration of XUV spectrometer, the spectra of harmonics are recorded when a 0.2-μm aluminum foil is placed in the XUV path, which can block the infrared (IR) beam and transmit the XUV beam. The filtered harmonic emission spectrum is shown in Fig. 1. The position of the aluminum absorption edge, which is 73 eV, can be clearly seen, as well as the discrete harmonics. This recorded spectrum shows very good resolution of XUV spectrometer.

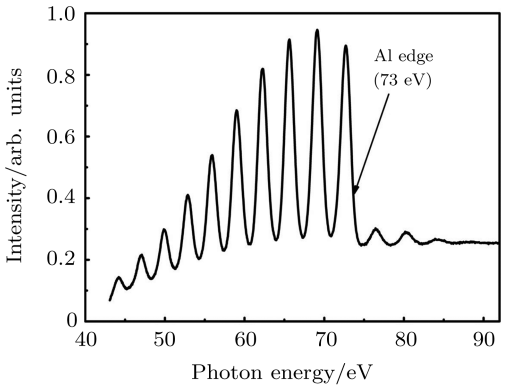

Fig. 1. Harmonic spectrum obtained by using a 0.2-μm aluminum foil, which is used for the calibration of spectrometer.

In order to compare the XUV spectrum distribution at cut-off region for the case of 25-fs and 5-fs laser pulse, the filter is changed to Zr foil with thickness of 0.2 μm which is used to transmit the XUV radiation. The transmission of Zr foil is shown in Fig. 2. The XUV spectra are obtained when the driver laser is 5 fs and 25 fs, as shown in Fig. 3. There are discrete and continuum spectra generated by 25-fs laser and 5-fs laser, respectively. In Fig. 3(a), the spectrum displays discrete spectral distribution, while when the driveing laser pulse is switched to 5 fs, the discrete harmonic spectra is smeared and becomes continuous distribution and the highest energy of HHG photon is extended to 131 eV, as shown in Fig. 3(b). These

different results obtained by 5-fs and 25-fs pulses can be explained as follows: during more than two periods of the optical pulse in the case of driving laser pulse of 25 fs, the electron reencounters the core more than two times, which results in the observation of relatively broad but still discrete harmonic spectral distribution, as described by simple semi-classical picture.[11,12] The continuous spectral distribution is unique to very short excitation pulses, for example 5-fs laser pulse in this case, harmonic generation occurs during very few reencounters of the electron with the core.[9,13] Due to ionization saturates at higher value of the laser field, when the driving laser pulse is much shorter (5-fs pulse), the spectral harmonics will extend to much higher energy at cut-off region, and is proved to be potential for generation of XUV source at water-window region.

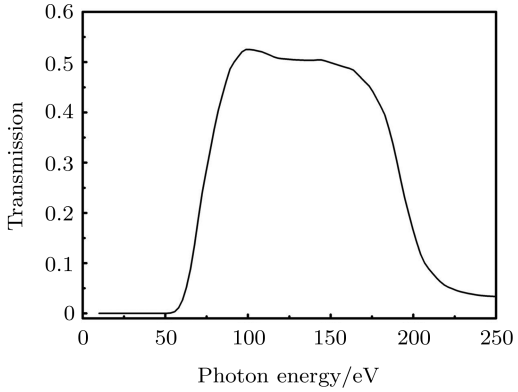

Fig. 2. The transmission of 0.2-μm Zr foil.

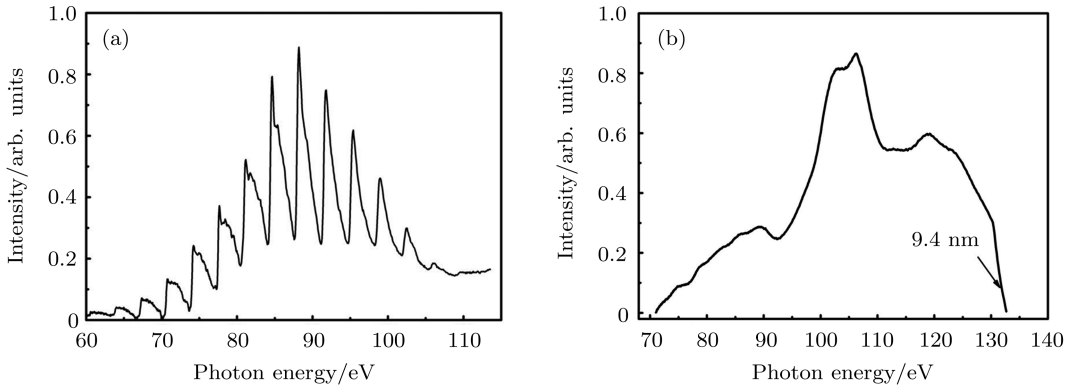

Fig. 3. Harmonic spectrum obtained from 25 fs and 5 fs respectively, when Zr foil with thickness of 0.2 μm is used as filter. The results show the discrete and continuum spectral distribution from (a) 25 fs and (b) 5 fs.

The continuum high order harmonics can be selected by using a simple broadband x-ray filter to produce isolated attosecond pulses, for these sufficiently short laser pulses the harmonic field generated during each reencounter only weakly depends on the history of the interaction (i.e. previous reencounter). Therefore there is no need to require a single-reencounter regime in order to generate attosecond pulses.[14] The dependence of HHG spectrum on CE phase is also very important for generation of single attosecond pulse, hence the emission of HHG can be controlled by the CE phase. The updated results are contained in Ref. [15].

4. Conclusions

The HHG spectral distribution between driving laser pulse of 25 fs and 5 fs is compared experimentally. Using 5-fs laser pulse as driving laser, we find that the harmonic emission becomes very broad spectrum, the highest orders almost merge together and become continuous distribution, the highest energy of coherent XUV radiation is extended to wavelengths of about 9 nm, whereas discrete spectral emission with lower energy XUV photon is presented when the driving laser pulse duration is 25 fs. This feature of XUV spectral emission by 5-fs laser pulse is due to the extremely short rising time of the driving pulses, neutral atoms can be exposed to higher fields before they are depleted by ionization. The intense XUV pulses delivered in a nearly diffraction-limited beam at kilohertz repetition rates offer the potential for attosecond resolution atomic spectroscopy and nonlinear optics in the XUV regime, this scheme is also proposed for generation of coherent x-ray in the water window.

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