Observation of non-odd order harmonics by sub-2-cycle laser pulses

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Abstract: High order harmonics generation from argon gas was comprehensively investigated by using multi-cycle and few-cycle laser pulses. Non-odd order harmonics were observed for sub-5-fs pulses, compare to the normal odd-order harmonics in the multi-cycle case. Theoretic analysis shows that the new spectral structure origins from the asymmetry of laser field in few-cycle pulses. This asymmetry induced both amplitude and phase difference between attosecond pulses from consecutive half-cycle of the laser field, which change the interference property of attosecond pulses and result in complex spectrum.

OCIS codes: (020.2649) Strong field laser physics; (190.4160) Multiharmonic generation; (320.7110) Ultrafast nonlinear optics.

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1. Introduction

High-order harmonic generation (HHG) from the extreme nonlinear interaction of ultra-short intense laser pulse with atoms or molecules has been intensively investigated since it opens the way of generating attosecond pulses, which is an ideal tool for studying the dynamic behavior of electrons in the atoms, molecules and surface [1,2]. The spectrum of HHG is consisted of odd- order harmonics of fundamental laser corresponding to an attosecond pulse train in the time domain. For attosecond pump probe experiments, single attosecond pulse is necessary. Single isolated attosecond pulsed can be generated using few-cycle laser pulse by limiting the generation process in a temporal duration with only one pulse included, such as amplitude gating [3], polarization gating [4] and double optical gating [5]. The generation of both odd and even order harmonics can be realized through the break of the symmetry of laser field by adding a static electric field [6], a weak second harmonic or high-order harmonics [7–9], or a mid-infrared laser field [10]. On the other hand, for HHG driven by the few-cycle pulses, the spectrum of the harmonics is very sensitive to the carrier-envelope phase (CEP) and components besides odd harmonics can be generated [11]. Mansten and associates [12] observed a complex spectral structure of harmonics from a two-color laser field with pulse duration of 12 fs, which can be used to determine the number of the attosecond pulses. However, the spectral signature of the structure of attosecond pulses is physically origined from the generation process, the two-color driven field is not essential. To avoid the perturbation of the second harmonic and confirm the physical origination of this spectrum structure, it is necessary to check this spectral signature again with one color few-cycle laser pulses.

In this paper, we studied HHG from argon gas driven by multi-cycle and sub-5-fs laser pulses respectively. Compare with the multi-cycle case, harmonic spectrum from sub-5-fs shows complex structure. Theoretical analysis shows this complex structure of the spectrum is attributed to the asymmetric of the laser field from few-cycle laser pulses.

2. Experimental setup and results

A standard Ti:Sapphire chirped-pulse amplifier (CPA) at a repetition rate of 1 kHz was used as the driving laser source, it deliveries pulse with energy of 1 mJ in 25 fs at the central wavelength of about 800 nm (Femtopower compact PRO, Femtolasers Produktions GmbH). The beam from this amplifier is focused into a gas filled hollow fiber with inner diameter of 250 μm to broaden the spectrum. In the experiment, neon gas is filled at the exit end of the hollow fiber and a vacuum pump connected to the entrance, so the pressure will gradually change along the fiber when the pump is running [13]. With this differential pump scheme, an ultra-broad spectrum covering the range from 420 nm to 960 nm is achieved at the pressure of 2.5 bar. Subsequently, a set of chirped mirrors plus a pair of wedges at small angle work as the compressor, with fine compensation of dispersion induced by air, gas and optical components for example optical window installed on the vacuum chamber, we obtained pulses with duration of about 4 fs and energy of 0.5 mJ. The second order interferometric autocorrelation trace of the pulse is shown in Fig. 1. For daily operation, a SHG interferometric autocorrelator is used as measurement of the duration of compressed pulses. Optimization of dispersion compensation by carefully tuning of wedge and gas pressure in hollow fiber, the laser normally works at sub-5-fs. By changing the pressure of gas in hollow fiber and the insert of wedge, the pulse duration can be lead to longer for example 15 fs, so we can easily change the pulse duration without any alignment of beam into the interaction chamber.

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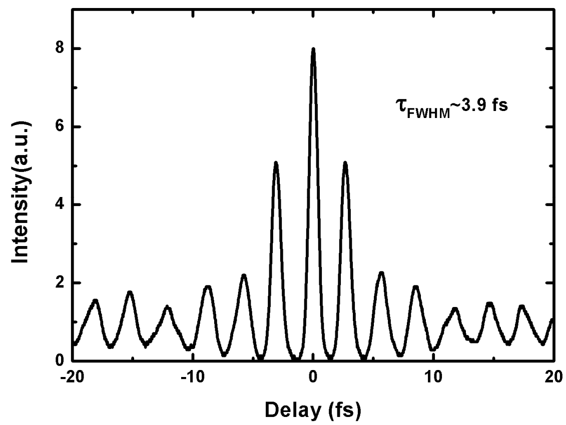

Fig. 1. The second order interferometric autocorrelation of compressed supercontinuum generated from differential pumped hollow fiber. The calculated pulse duration is 3.9 fs (FWHM), corresponding to 1.5-cycle with a carrier wavelength of 780nm

To obtain the HHG emission, this sub-5-fs laser pulses are gently focused into a quasi-static gas cell filled with argon gas at a pressure of ~100 mbar. The laser beam is focused by a silver- coated spherical mirror with focal length of 35 cm through a 1 mm- thickness fused silica window with high transmission broadband coating into a vacuum chamber. The pulse energy is ~0.33 mJ on the target, and is focused to a $1/e^2$ diameter of ~60 µm, corresponding to the intensity is about $\sim 10^{15}$ W/cm^2 on the beam axis. The HHG radiation produced collinearly with the laser beam was sent through a 1- mm aperture into a grazing incidence flat-field spectrometer evaluated to $< 10^{-4}$ mbar. The flat-field spectrometer is equipped with 1200 lines/ mm (mean value) varied line-spacing concave grating and soft X-ray CCD (PIXIS- XO, Princeton Co.). To block the fundamental laser light completely, a 200 nm-thickness aluminum foil with transmission from 60 nm to 16.9 nm is inserted before the slit in the spectrometer. The experimental setup [14] is shown in Fig. 2. The focus point of laser beam is located about 5 mm before the gas target to ensure the short electron trajectories being dominated the harmonic signal [15].

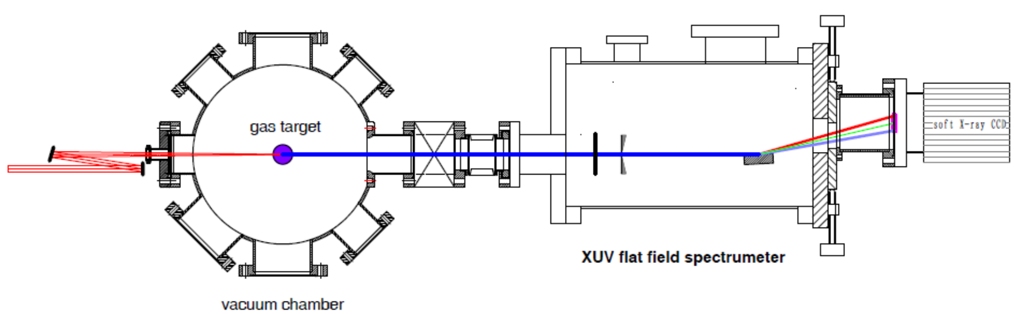

Fig. 2. Experimental setup for generation and measurement of HHG spectrum

Based on above experimental system, HHG spectrum driving by multi-cycle 15 fs and sub-5-fs laser pulses are recorded respectively, which is shown in Fig. 3. Figure 3(a) shows the HHG spectrum driving by sub-5-fs, and the HHG spectrum driving by 15 fs is shown in Fig. 3(b). The sharp edge at 16.9 nm in the two spectrums is the absorption edge of aluminum. Generally, a broad continue spectrum close to the cutoff exist for sub-5-fs case, which represent a single attosecond pulse. But in this work we did not focus on the cut-off

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region and attosecond pulse, we are focusing here is the region between the plateau and the cutoff, the spectrum in this region is still modulated or discrete distribution. As we can see in Fig. 3, a typical HHG spectrum with only odd-order harmonics and discrete distribution is obtained when the driving laser pulse is 15 fs. When the pulse's duration is switched to sub-5-fs, some new features of HHG spectrum occurs: The spectrum of each order become more broader, and peak of harmonic present red-shifted, more interesting is that additional components appear between the consecutive odd orders, and results in complex structure distribution.

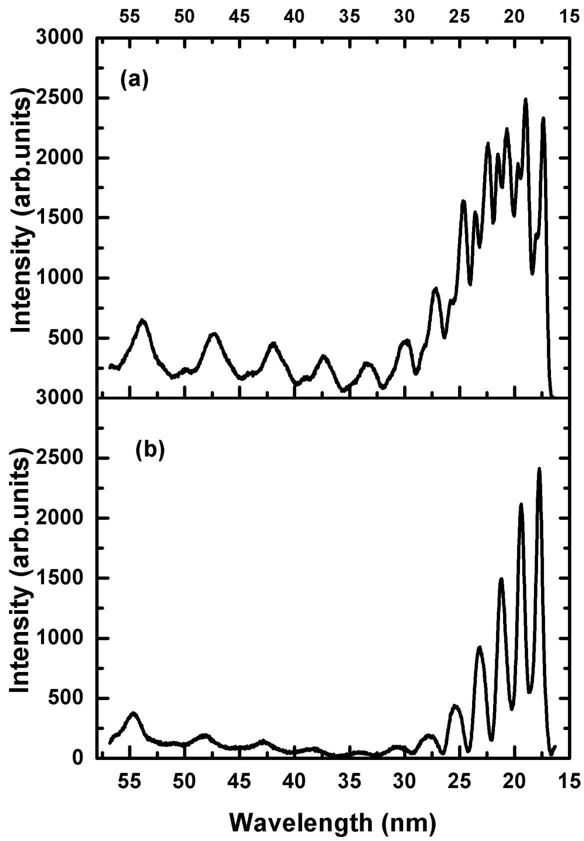

Fig. 3. High-order harmonic spectrum from argon gas show complex structure distribution of spectrum with additional frequency components are located between the consecutive odd-order harmonics when driving laser pulses is only sub-5-fs (a), and discrete odd-order HHG distribution when driving laser pulses consist of multi-cycle (b)

3. Discussion

The standard HHG spectrum distribution with odd-order from multi-cycle laser pulses in Fig. 3 is well understood. The HHG emission occurs at every half-cycle of the fundamental which corresponding to 2ω spacing of consecutive spectrum, and present odd-order harmonics of the driver laser. The emission can be expressed as:

$$S(t)=\sum_{j=1}^{N}a_{+}(t)\otimes\delta(t-jT)-\sum_{j=1}^{M}a_{-}(t)\otimes\delta(t-jT-T/2) \quad (1)$$

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Where $a_+(t)$ and $a_-(t)$ are the amplitude of the attosecond pulses emitted in the first '+' and second '-' half-cycle of the driving laser field, T is the cycle of laser field (*T=2.7fs* for our experiment). We assume that $a_+(t) = a_-(t) = a(t)$, which means that the attosecond pulses from the different cycle of laser field has the same amplitude, the Fourier transform of the pulse train can then be approximated as:

$$S(\Omega) \approx A(\Omega)(\sum_{j=1}^{N} e^{ij\Omega T} - \sum_{j=1}^{M} e^{ij\Omega T + i\frac{\Omega T}{2}}) \tag{2}$$

Here, $S(\Omega)$ and $A(\Omega)$ are the Fourier transform of $s(t)$ and $a(t)$ respectively. For multi-cycle laser pulses, the electric field for positive and negative directions are symmetric, $M = N$. We can conclude that $S(\Omega) = 0$ for $\Omega = 2n$ ($n = 1, 2, 3...$), which means the even order harmonics disappear and only odd-order harmonics exist for multi-cycle laser pulses. But for sub-5-fs laser pulses, which consist of field oscillation less than 2 cycles, the electric field is spatial asymmetric and this asymmetry is strongly dependent on the CEP. For different CEP of sub-2-cycle laser pulses, except for CEP = π/2, $M \neq N$, then $S(\Omega) \neq 0$, which means both even and odd order harmonics exist.

The analysis above show that the spatial distribution of laser field for positive and negative directions is so asymmetric for this sub-2-cycle laser pulse that the number of attosecond pulses from consecutive positive and negative direction of laser field oscillation is different, the even orders cannot be completely canceled through the interference. Actually the electric field of the few-cycle laser pulse is also completely temporal asymmetric, the amplitude of the field changes drastically between successive half optical cycle, so $|a_+(t)|$ and $|a_-(t)|$ are not exactly the same. Also this result in substantial difference on the relative phase between the neighbor attosecond pulses, because one of the major part of harmonic phase comes from the phase accumulation of electron in the continuous states, which is strongly proportional to the driving laser intensity. All these differences change the temporal interference between the attosecond pulses, result in a complex spectral structure in frequency domain. From this analysis, the complexity of the harmonic spectrum structure distribution is attributed to the spatial and temporal asymmetry of the laser field of sub-2-cycle pulses. The analysis is corresponding to the experiment results which show complex structure distribution of HHG when the driving laser pulse is sub-5-fs.

4. Summary

High order harmonics generated from argon gas driving by multi-cycle and few-cycle laser pulses have been studied comprehensively. Complex spectral structure for sub-5-fs is observed compare with normal odd order harmonics in the multi-cycle case. Theoretic analysis shows this spectral structure origins from the asymmetry of laser field in few-cycle pulses. This asymmetry induced both amplitude and phase difference between attosecond pulses from consecutive half-cycle of the laser field, which change the interference property of attosecond pulses and result in complex spectrum.

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