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Complex Spectra Structure of an Attosecond Pulse Train Driven by Sub-5-fs Laser Pulses *

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We present the observation of the additional spectral components between the odd order harmonics in the harmonic spectrum generated from argon gas driven by sub-5-fs laser pulses. The theoretical analysis shows that the asymmetric laser field in both spatial and temporal domains leads to this complicated spectrum structure of high order harmonics.

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Extreme ultra-violet (XUV) sources have extensive applications in the ultrafast spectroscopy of materials,[1,2] in the analysis of structures,[3] as well as in high resolution imaging. To produce XUV emission, high harmonic generation is one of the best methods,[4] which is implemented through an extreme nonlinear interaction between intense laser pulses and noble gases.[5,6] High harmonic generation (HHG) is a re-collision process of the atomic core with field ionized electrons, which are driven away and then returned by the oscillating field when the atom is exposed to an intense laser field.[7,8] The re-collision occurs twice in one oscillation of the laser field, corresponding to 2ω spacing in the frequency domain, which presents odd order harmonics of the driver laser, due to the symmetry of each oscillation in ideal driving laser pulses consisting of many cycles,[9−11] for example, 50 fs. When the symmetry of the laser field in each oscillation is broken, this will lead to only one re-collision event in the full cycle of the laser field and will result in both even and odd order harmonics generation. The break of the laser field oscillation symmetry can be realized by the methods as follows: one is a fundamental laser field adding a weak second harmonic field,[12] the other is a laser field adding a static electric field.[13] Many experimental results of a two-color laser field show observation of even order harmonic generated.[14−16] For example, Mansten and co-workers[17] observed a complex structure in the spectral region close to cutoff from a two-color laser field with pulse duration of 12 fs. However, for the sub-2-cycle laser field in our case, it is essentially asymmetry, and it is interesting which half of the cycle field peak in the centre is more intense than the other for cosine or sine waveform, so the asymmetry of the laser field results in a break in the inversion symmetry, and even order harmonics can be generated.

In this study, we observe the additional spectral components between the odd order harmonics, generated from argon gas driven by sub-5-fs laser pulses, which shows a complex structure in the HHG spectrum. The asymmetric laser field of the sub-2-cycle pulses in both spatial and temporal domains leads to this complicated spectrum structure of high order harmonics.

A standard Ti:sapphire chirped pulse amplifier (CPA) at a repetition rate of 1 kHz was used as the driving laser source. It delivers a 25 fs laser pulse with energy of 0.8 mJ at the central wavelength of about 800 nm. The output laser pulses are focused into a neon filled hollow fiber to broaden the spectrum with a differential pump scheme. Subsequently, there are a set of chirped mirrors and a pair of wedges at small angles working as the compressor. A sub-5-fs laser with a pulse energy of 0.5 mJ is obtained. The details of laser performance have been described in Ref. [18].

To obtain HHG radiation, the sub-5-fs laser pulses were gently focused by a silver-coated spherical mirror with a focal length of 35 cm into a quasi-static gas cell, which was filled with argon gas at a pressure of about 100 mbar. A 1-mm-thick fused silica window with a high transmission broadband coating was used for the vacuum chamber, which contains the gas target at the focus. The pulse energy is about 0.33 mJ on the target and is focused to a $1/e^2$ diameter of about 60 μm, corresponding to an intensity of about 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 with a vacuum evaluated to be $<10^{-4}$ mbar. The flat-

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field spectrometer is equipped with a 1200 line/mm (mean value) varied line-spacing concave grating and soft-x-ray CCD (PIXIS-XO, Princeton Co.). To block the fundamental laser completely, a 200-nm-thick aluminum foil with a transmission band from 16.9 nm to 6 nm was inserted before the slit in the spectrometer. The experimental setup is shown in Fig. 1.

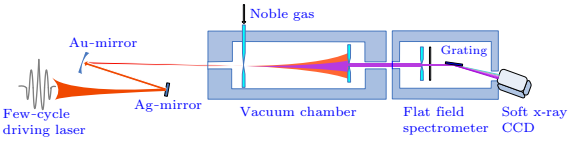

Fig. 1. Experimental setup for the generation and measurement of the HHG spectrum.

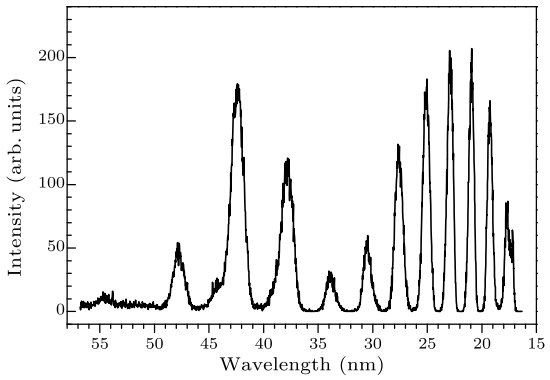

Fig. 2. A high-order harmonic spectrum from Ar gas when driving laser pulses consists of multi-cycles, which show the discrete odd-order HHG distribution.

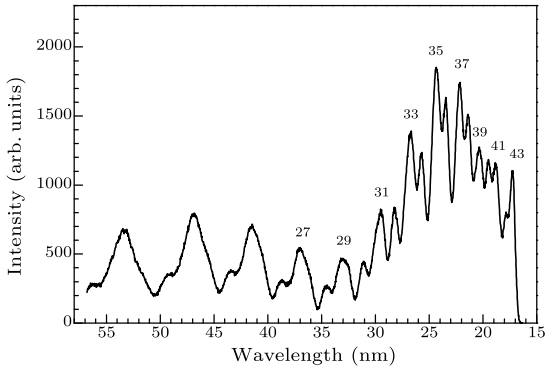

Fig. 3. High-order harmonic spectrum from Ar gas when the driving laser pulses are only sub-2-cycle pulses, which show a complex structure distribution of spectrum with extra frequency components located between consecutive odd order harmonics.

Based on the above experimental system, we investigated the asymmetry of the sub-2-cycle laser field from the complex structure of the HHG emission spectrum. We obtained the HHG spectrum driven by multi-cycle 15 fs and sub-2-cycle laser pulses, respectively, as shown in Figs. 2 and 3. In these two figures, the sharp decrease at 16.9 nm in the spectrum is the absorption edge of the aluminum filter. Figure 2 shows the typical HHG spectrum with only odd order harmonics and discrete distribution when the driving laser pulse is 15 fs. When the laser pulse duration switches to sub-5-fs, the HHG spectrum structure changes a lot. There are extra components between the odd orders, resulting in a complex structure distribution shown in Fig. 3.

The normal HHG spectrum distribution with odd order in Fig. 2 is well understood. The HHG emission occurs at every half cycle, which corresponds to 2ω spacing of the consecutive spectrum of HHG, and there appear to be odd-order harmonics of the driver laser. The emission is expressed as

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

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 ($T = 2.7$ fs in our experiment), respectively. N and M represent the numbers of emission. We assume that $a_+(t) = -a_-(t) = a(t)$, which means that the attosecond pulses from different half cycles of the laser field have the same amplitude. The Fourier transform of the pulse train can then be approximated as

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

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

The above analysis shows that the spatial distribution of the laser field is so asymmetric for the sub-2-cycle laser pulse that the numbers of attosecond pulses from consecutive positive and negative electric field directions are different, and the even orders cannot be completely canceled through the interference. Actually, the electric field of the few-cycle laser pulse is also completely asymmetric in the time domain, the amplitude of the field changes a lot within the successive

half optical cycle, so $|a_+(t)|$ and $|a_-(t)|$ are not exactly the same. Also, this results in a substantial difference in the relative phase between the neighbor attosecond pulses, because one major part of the harmonic phase comes from the phase accumulation of electrons in the continue states, which is strongly proportional to the driving laser intensity. All these differences change the temporal interference between attosecond pulses in the train, resulting in a complex spectral structure in the frequency domain. From this analysis, the complexity of the harmonic spectrum structure distribution is attributed to the spatial and temporal asymmetry of the sub-2-cycle laser pulses. The analysis corresponds to the experimental results, which show complex structure distribution of HHG when the driving laser pulse is sub-2-cycle.

In summary, we have investigated the dependence of a high harmonic spectrum on the asymmetry of the laser field when the driving laser pulses consist of only sub-2-cycle pulses. The complex structure distribution of the HHG spectrum is obtained, which is compared to the standard discrete odd order HHG spectrum for driving laser pulses with duration of multicycle. The theoretical analysis shows that both the spatial and temporal asymmetry of this sub-2-cycle laser pulse leads to the complexity of the harmonic spectrum structure distribution.

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