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Generation of Continuum Extreme-Ultraviolet Radiation by Carrier-Envelope-Phase-Stabilized 5-fs Laser Pulses *

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Coherent extreme-ultraviolet (XUV) radiation is studied by interaction of carrier-envelope (CE) phase stabilized high energy 5-fs infrared (800 nm) laser pulses with neon gas at a repetition rate of 1 kHz. A broadband continuum XUV spectrum in the cut-off region is demonstrated when the CE phase is shifted to about zero, rather than modulated spectral harmonics when setting of CE phase is nonzero. The results show the generation of isolated attosecond XUV pulses.

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The fundamental processes of chemistry, biology, and material science are mediated by the electronic and nuclear motions of the constituent atoms. The extremely fast electronic motions inherent to these systems have an attosecond time scale (10^{-18} s). Attosecond metrology has already been demonstrated as a viable means for time-resolving the fastest electronic processes.[1] Generation of isolated attosecond pulses is crucial to the development of these techniques.

There are two processes that can generate attosecond pulses: high order harmonic generation (HHG) from atoms exposed to intense optical pulses[2] and multiple-order Raman side band generation in molecular media.[3] Compared to the multiple-order Raman side band generation technique, the high harmonic generation in noble gases is a powerful mean of generation of the single attosecond XUV pulse, due to its providing coherent XUV radiation from a laser-driven source.[4,5,18,19] With the advent of intense few-cycle laser near-infrared (NIR) and its wave-form controlled technique,[6] the isolation of attosecond or sub-femtosecond XUV light bursts becomes feasible.[7,8] The key breakthrough has shown in Ref. [9] that the XUV pulses as short as 500 are demonstrated based on high harmonic generation by carrier-envelope (CE) phase-controlled optical pulses. With these techniques, the reproducible production of the isolated XUV attosecond pulses with a pulse duration of 250 as at about 100 eV[8] and 130 as at 35 eV,[11] 170 as at 90 eV,[10] and as shot as 80 as at 80 eV[12] are obtained. With these isolated attoseocnd XUV pulses, scientists have demonstrated some time-resolved spectroscopy experimental studies on the temporal evolution of atomic scale electron motions[13] and secondary electrons (Auger) emission.[14]

In this Letter, we report a continuous spectral range down to sub-10 nm driven by CE-phase-controlled 5 fs laser pulses into a neon gas cell. When the phase is shifted to about zero, the continuum spectra in the cut-off region is obtained, which corresponds to the isolated attosecond XUV pulses.

A Ti:sapphire chirped-pulses amplifier delivers 25 fs with energy of 1 mJ at a repetition rate of 1 kHz. By focusing the pulses from the amplifier into a hollow fiber filled with neon gas, spectrally broadened pulses are re-compressed down to 5-fs duration with chirped-mirrors. These quasi-monocycle laser pulses contain 0.5 mJ, and have nearly diffraction-limited beam quality.[17] Because the CE-phase is key for the generation of the isolated XUV pulse, the CE phase must be controlled. The CE phase is controlled by utilizing two feed-back control loops, one is a fast loop based on difference-frequency generation in PPLN crystal, an interferometric beat signal can be detected and sent to a fast feedback loop, in which the pump power is modulated by an acousto-optical modulator (AOM). The phase controlled laser pulses from oscillator serve as seed for the Ti:sapphire chirped-pulse amplifier (CPA) system, the slow CE-phase drifts imparted to the amplifier are compensated for by an $f-2f$ spectral interferometer installed after the compressor. Based on these two CE-phase stabilization schemes, the resulting phase stability of sub-5-fs pulses was on the order of about 50 mrad rms over several hours.[15,16]

Coherent extreme ultraviolet (XVU) light is generated by gently focusing the 5-fs laser pulses into a quasi-static gas cell filled with neon gas at a pressure of about 200 mbar. The laser beam is focused with

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a silver-coated spherical mirror with a focal length of 35 cm through a 1-mm-thick fused silica window with a high transmission broadband coating into a vacuum chamber which contains the 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, the correspondingintensity is about about 10^{15} W/cm^2 on the beam axis. The XUV radiation produced collinearly with the laser beam was passed 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 laser light completely, a 200-nm-thick zirconium foil is inserted before the slit in the spectrometer. The experimental setup is shown in Fig. 1.

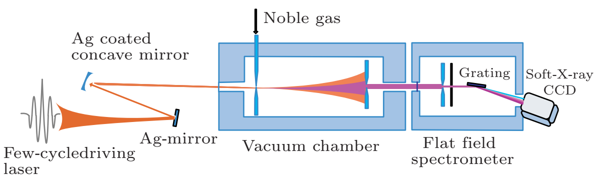

Fig. 1. Experimental setup for generation and measurement of XUV spectrum.

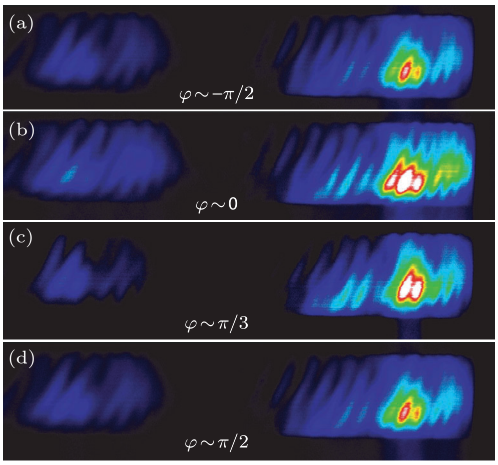

Fig. 2. Harmonic spectrum images obtained when the CE-phase is changed.

When the driving laser pulse is much shorter, generated harmonics peaks will also be broadened, which will result in the consecutive peaks of harmonics merging to a continuum. To rule out this possibility that the continuum is caused by the accumulation of many peak broadened spectra, the CE-phase is stabilized and the focus is located at 5 mm before the gas cell to ensure that short electron trajectories dominate the harmonic signal.[11]

To investigate the dependence of the resultant XUV spectrum on the CE-phase of driving laser pulses, XUV spectra were recorded for different settings of the CE-phase, as shown in Figs. 2 and 3. Figures 2(a)–2(d) show a series of typical XUV spectrum image recorded at different CE phases of laser pulses, the CE phases are taken from $-\pi/2$ to $\pi/2$, stepped in increments of $\pi/4$. From Fig. 2, notably, with a change of the CE phase, the cut-off XUV spectrum gradually transform from discrete modulated harmonic peaks to continuum spectral distribution. Figure 2(b) shows a broad structureless continuum spectrum appearing in the cut- off region when the CE phase is close to zero. As the CE-phase is slowly varied between zero and π by inserting a wedge in optical path, the continuum spectrum in the cut-off region becomes much modulated and shows discrete harmonic peaks, and this modulation will become maximal when the phase is equal to $\pm\pi/2$. This behavior is periodic and observed to repeat upon subsequent full π phase shift.

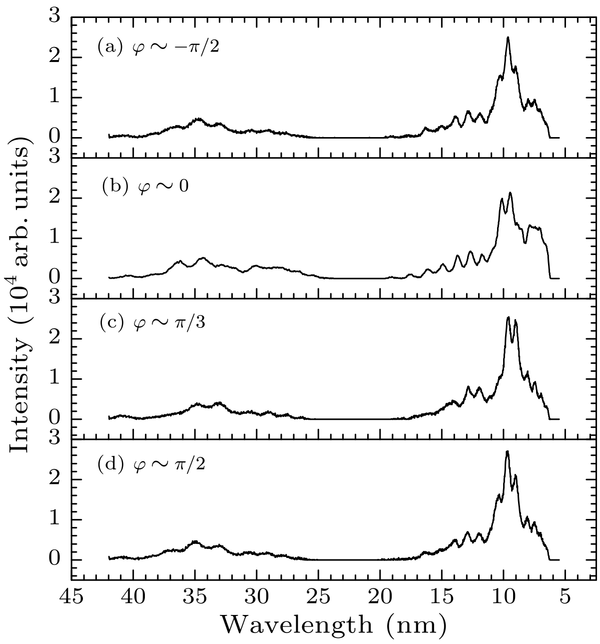

Fig. 3. Lineout of spectrum in Fig. 2, (b) the spectrum in the cut-off region from continuum distribution when the phase is close to zero, and the generation of the single attosecond pulse.

The continuum XUV spectrum in the cut-off region is corresponding to the theoretical studies,[7] for a Cosine waveform (CE phase of about 0), there is only one single highest energy XUV burst to be emitted, which forms a continuum cut-off spectral, whereas the sine waveform (CE phase of about $\pi/2$), comprising two or three half cycles of the highest amplitude, will create a couple of XUV pulses at the highest photon energies. The continuum spectrum can be selected by filter, which corresponds to isolated attosecond pulses. In Fig. 3(b), the spectrum in the cut-off region forms a continuum distribution which covers 1.5 nm at a central wavelength of 7.5 nm. From the continuum distribution spectrum, the temporal profile can be derived from the Fourier transform assuming flat phase, as shown in Fig. 4, the continua spectrum is capable of supporting Fourier-transform-limited pulses with pulse duration of about 100 as.

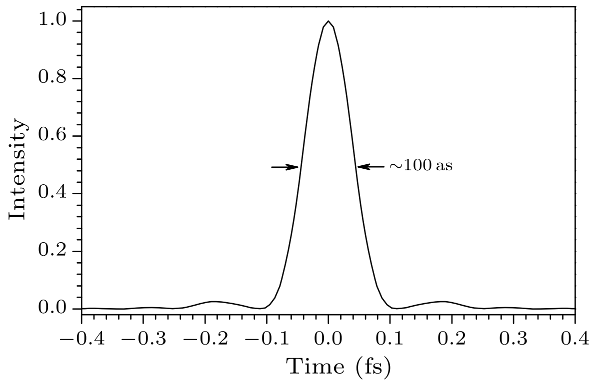

Fig. 4. Temporal profile derived from the Fourier-transform assuming flat phase, which is in agreement with the continuum spectrum distribution in Fig. 3(b). The profile shows that the spectrum is capable of supporting about 100 as.

In conclusion, we have studied the dependence of the XUV spectrum on the CE-phase of 5-fs laser pulses, XUV spectra are recorded for different settings of the CE phase. The continuum XUV spectrum in the cut-off region has been demonstrated when the CE-phase is shifted to zero, in which the continuous XUV spectrum has a bandwidth of about 1.5 nm at the central wavelength of 7.5 nm. The results show the generation of isolated attosecond XUV pulses.

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