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2013 Chinese Phys. Lett. 30 093201

(http://iopscience.iop.org/0256-307X/30/9/093201)

CHIN. PHYS. LETT. Vol. 30, No. 9 (2013) 093201

Generation and Measurement of Isolated 160-Attosecond XUV Laser Pulses at 82 eV *

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(Received 17 April 2013)

Isolated attosecond extreme-ultraviolate (XUV) pulses are generated based on high-harmonic-generation from a neon gas cell driven by carrier-envelope phase stabilized sub-5-fs Ti:sapphire laser pulses at repetition rate of 1 kHz. Temporal characterization of isolated attosecond XUV pulses is demonstrated to be 160-attosecond by attosecond streaking spectroscopy. The development of attosecond source and streaking spectroscopy will allow scientists to explore the electron dynamics in matter.

PACS: 32.30.Rj, 42.65.Re, 32.80.Fb, 42.55.Vc DOI: 10.1088/0256-307X/30/9/093201

With the advent of isolated attosecond XUV pulses (IAPs) ten years ago,[1] scientists can gain insight into the electronic motion such as charge transfer, atomic inner shell transitions. Traditional femtosecond pump-probe technique cannot access the time domain of sub-femtosecond that electronic processes naturally have. The development of attosecond source and adequate time resolved technique provides access to this time regime to realize direct measurement of electron dynamics in chemical reactions, the study of electronic motion in strong field, and the exploration of dynamics of inner shell excitations in matter.[2]

So far, several methods have been developed for generation of IAPs. The first method is amplitude gating,[3] which selects a continuous cutoff region in the spectrum of high-harmonic-generation driven by carrier-envelope phase (CEP) stabilized few-cycle laser pulses. IAPs as short as 80-attoseconds are obtained by this method.[4] The second approach is polarization gating,[5] which creates a pulse with time varying polarization sweeping from circular through linear back to circular and forms a quite short window for generation of IAPs. Nisoli's group employed polarization gating technique with CEP stabilized 5-fs laser pulses to produce 130-as pulses,[6] and further development of above-saturation few-cycle laser fields generates higher energy sub-160-as IAPs.[7] The third method is ionization gating, which provides temporal gating for phase mismatching between the fundamental and harmonic fields. There are 430-as pulses generated by this method.[8] Recently, double optical gating (DOG)[9] and general double optical gating (GDOG),[10] which are a combination of polarization gating and two-color gating, were developed. These schemes relax the requirement on the pulse duration to 28 fs, making it easier to generate IAPs and to scale to higher energy IAPs. Using 7-fs driving laser, a much broader continuous XUV spectrum covering both plateau and cutoff region is generated and demonstrated to be the shortest 67-as based on DOG.[11]

In parallel with the development of IAPs' generation, remarkable progress in attosecond metrology has been achieved. Due to the limitation of low energy, attosecond pump-probe spectroscopy cannot be realized. Alternatively, attosecond streaking spectroscopy employs IAPs as pump and femtosecond IR laser pulses as probe or vice versa.[12,13] The electrons emitted from atoms by an impulse excitation of attosecond XUV pulses experience acceleration by the electric field of the IR laser pulse. The final kinetic energy of accelerated electrons can be recorded by a time-of-flight (TOF) spectrometer. By scanning the delay between the attosecond XUV pulse and femtosecond IR pulse, the attosecond streaking spectrogram can be obtained. The characterization of attosecond pulses can be obtained from the spectrogram by FROG for complete reconstruction for attosecond burst (FROG-CRAB)[14] or PROOF.[15] The attosecond streaking spectroscopy can also be applied for attosecond-resolution studies, due to the laser-induced energy shift of electrons which results in a unique mapping of the temporal emission profile into a similar energy distribution. Probing the photoelectrons or Auger electrons emitted by XUV burst with this pump-probe technique allows real time observation of atomic excitation and subsequent relaxation processes with attosecond resolution.[16–18] The at-

*Supported by the National Basic Research Program of China (973 Program) under Grant No 2013CB922402, the Instrument Developing Project of the Chinese Academy of Sciences under Grant No 2010004, and the National Natural Science Foundation of China under Grant No 11074298.

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tosecond streaking spectroscopy can further characterize the waveform of femtosecond IR laser pulses; the attosecond photoelectron burst creates a probe for tracking variations of the electric field of an electromagnetic wave with attosecond resolution as reported in Refs. [19,20]. Most recently, this technique realizes the observation of optical-field-induced current in dielectrics[21] and control of dielectrics with the electric field of light.[22]

In this Letter, we report that IAPs are generated based on high-harmonic-generation from neon gas cell driven by CEP stabilized sub-5-fs laser pulses at repetition rate of 1 kHz. The temporal characterization of IAPs is demonstrated to be 160-attosecond at 82 eV by homemade attosecond streaking spectroscopy.

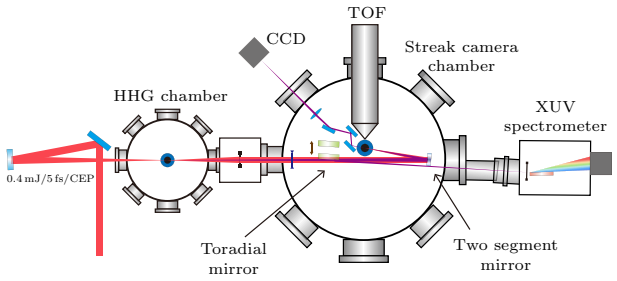

Fig. 1. The schematic of the attosecond beamline, which consists of the CEP stabilized sub-5-fs laser, HHG chamber, attosecond streaking spectroscopy chamber and XUV spectrometer.

The experimental setup consists of CEP stabilized sub-5-fs Ti:sapphire laser, high-harmonic-generation chamber, attosecond metrology chamber, and flat-field XUV spectrometer, as shown in Fig. 1. The driving laser for generation of IAPs is a CEP stabilized sub-5-fs Ti:sapphire laser which is achieved by spectral broadening in neon-filled hollow fiber and chirped mirror compressor. The shortest pulse duration of less than 5-fs with energy of 0.5 mJ is obtained. The detail of the laser system is described in reference.[23] The CEP is locked by utilizing two separated feedback control loops, one is the fast control loop for stabilization of laser pulses from the oscillator, the other one is the slow loop for stabilization of pulses from amplifier by controlling the displacement of one prism in the compressor based on the feedback signal of f–$2f$ spectral interferometer installed after the hollow fiber. Based on the two CEP stabilization schemes, phase stability of sub-5-fs pulses is of the order of ∼60 mrad (rms) over 3 h.

Attosecond XUV pulses are generated by focusing the driving laser pulses into a quasi-static gas cell filled with neon gas at a pressure of ∼200 mbar. The laser beam is focused by a silver-coated spherical mirror with focal length of 500 mm through a 1 mm fused silica window with high transmission broadband coating into a vacuum chamber which contains the gas cell at the focus. The focal intensity is $\sim 8 \times 10^{14}$ W/cm^2 on the beam axis. XUV radiation propagates collinearly with the IR beam through a motorized aperture into the attosecond streaking spectroscopy chamber for characterization of attosecond pulses. The attosecond streaking spectroscopy chamber consists of neon gas target, TOF spectrometer, two-segment-mirror delay line which is assembled with inner Mo/Si multilayer mirror and annular silver-coated mirror. A 150-nm-thick Zr foil mounted at the center of a 5-μm-thick pellicle acts as a filter for transmission of the attosecond XUV beam and block of the IR beam. The IR beam transmits through the pellicle in the form of an annular beam. These two concentric beams (XUV+IR) are focused by the two-segment-mirror with the same focal length of 125 mm. The inner Mo/Si mirror is used for reflecting XUV beam, while the outer silver annual mirror is used for reflecting IR beam. The delay between XUV and IR pulses is adjusted accurately by translating the inner mirror along the axis on a piezo stage. With this scheme, the IAPs are focused together with IR pulses overlapped spatially and temporally into the second neon gas target. The attosecond XUV pulses firstly ionize neon to generate photoelectrons, and then the photoelectrons are shifted in momentum by an amount that depends on the vector potential of IR field at the time when photoelectrons are emitted. By scanning the delay between the XUV and IR pulses, the modulated photoelectron momentum at each delay time creates an attosecond streaking spectrogram.

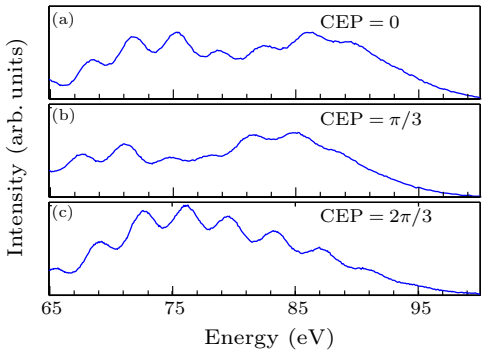

Fig. 2. Spectrum of high-harmonic-generation with dependence of CEP.

By changing the CEP of sub-5-fs laser pulses through adjusting an inserted wedge in the optical beam after the hollow fiber, the cutoff harmonic spectrum gradually transforms from a structure with discrete modulated harmonic peaks to continuum distribution, as shown in Fig. 2. This behavior is periodic and observed to repeat upon subsequent full π phase shift. When the CEP is close to 0, which corresponds to cosine waveform, the spectrum shows discrete harmonic peaks in the plateau region but a continuous spectrum in the cut-off region which supports generation of IAPs. The characterization of IAPs is per-

formed in attosecond streaking spectroscopy. The attosecond streaking spectrogram is shown in Fig. 3(a). The delay is scanned over a range of 16 fs in steps of 100-as and the TOF signal of photoelectrons is integrated for 2000 pulses. The obtained photoelectrons are centered at the energy of 82 eV with 15 eV shift of the electron peak. The strong oscillation of the photoelectron energy is observed in the region of temporal and spatial overlap. The results also demonstrate that the IR pulse is CEP stabilized.

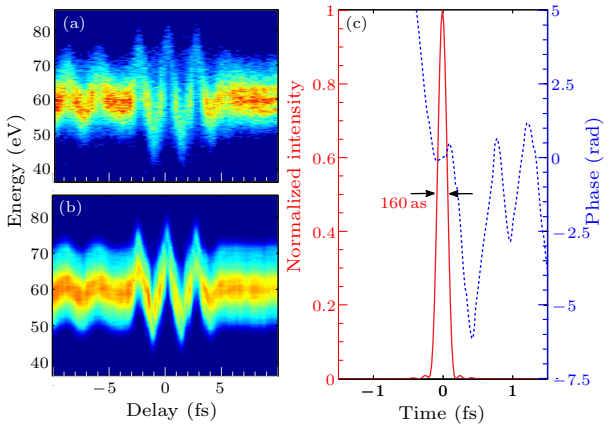

Fig. 3. (a) Attosecond streaking spectrograms delay between the XUV and IR pulses, (b) FROG-CRAB reconstruction, and (c) retrieval of temporal amplitude and phase of IAPs.

The retrieval of the duration of IAPs is carried out by FROG-CRAB algorithm. The attosecond streaking spectrogram, proportional to the transition probability to the final state due to photoelectrons ionized by an XUV pulse in the presence of IR laser field shifted by a variable delay, is given by

$$I(p,\tau) = \left| \int_{-\infty}^{\infty} E_X(t) e^{i\phi_L(t+\tau)} e^{i(p^2/2+W)t} dt \right|^2, \quad (1)$$

$$\varphi_L(t+\tau) = -\int_{t+\tau}^{\infty} (p_c A_L(t') - \frac{1}{2} A_L^2(t')) dt', \quad (2)$$

where p is the instantaneous momentum of electrons, τ is the delay between XUV and IR, E_X is the electric field of the XUV, P_c is the central momentum of electrons emitted by XUV, A_L is the vector potential related to electric field of IR by $E_L(t) = -\partial A_L/\partial t$, W is the ionization potential, and $\Phi_L(t)$ is the phase gate induced by IR field.

FROG-CRAB can iteratively converge to give the electric field of the XUV pulse and IR pulse. The algorithm is applied to the streaking spectrogram obtained, and gives the retrieved XUV pulse duration of 160-as shown in Fig. 3(c). As described above, the electric field of IR pulses can also be reconstructed from the same spectrogram. The electric field of the femtosecond IR pulse reconstructed is shown in Fig. 4, the circle is experimental data from the FROG-CRAB algorithm and the solid line is fitting curve, the error at the peak of electric field is due to the lower count number at maximum energy shift, the reconstructed electric field with the duration of less than two-optical cycle is consistent with our previous autocorrelation measurement.[23]

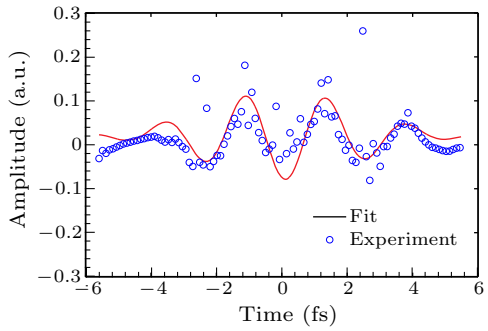

Fig. 4. The electric field of IR pulse reconstructed from the attosecond streaking spectrogram. The circle is experimental data, solid line is fitting curve.

In conclusion, the IAPs are successfully generated based on high harmonic driven by the CEP stabilized sub-5-fs laser pulses at repetition rate of 1 kHz. The temporal characterization of attosecond pulses is carried out based on attosecond streaking spectroscopy, and the results show that the beamline can deliver the IAPs with pulse duration of 160-attosecond at the central wavelength of 82 eV. With the development of attosecond source generation and attosecond streaking spectroscopy, the XUV source with attosecond resolution will open new applications in condensed matter physics.

We thank F. Krausz, Zenghu Chang, J. W. G. Tisch for helpful discussion.

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