JOURNAL OF MODERN OPTICS, 20 JANUARY–15 FEBRUARY 2005
VOL. 52, NO. 2–3, 277–283

Generation of the attosecond extreme ultraviolet supercontinuum by a polarization gating

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(*Received 18 April 2004; revision received 22 May 2004*)

Abstract. We report the first demonstration of extreme ultraviolet (XUV) supercontinuum generation in the plateau region of the high-order harmonic spectrum indicative of a single attosecond pulse. It was accomplished by combining the generation of sub-10 fs laser pulses with the polarization gating of high harmonic generation. When an 8 fs pulse centred at 750 nm was split and delayed with a quartz plate and then recombined with a quarter waveplate, a laser pulse was created whose polarization varied rapidly from circular to linear and back to circular. The near-linearly polarized portion of the resultant pulse was only 1.3 fs long, which was much shorter than the laser pulse duration. Since the high-order harmonic generation process is susceptible to the ellipticity of the driving field, the pulse with a time-dependent ellipticity behaved like a half-cycle linearly polarized pulse for generating XUV radiation. By exciting argon gas with the pulse, a supercontinuum that covered 25–45 nm was produced, which corresponds to an estimated single 200 as pulse.

1. Introduction

High-order harmonic generation (HHG) has been proved to be an effective process for generating both a train of attosecond pulses and single isolated attosecond pulses [1–3]. It is now well accepted to describe the HHG by a three-step semiclassical model. The model is valid in the tunnelling regime of laser–atom interaction. Under an intense, linearly polarized laser field, the valence electron in an atom or molecule tunnels through the Coulomb barrier. The freed electron has a certain probability to be driven back to its parent ion and to recombine with it to generate an attosecond burst of electromagnetic waves. This process occurs every half-cycle of the laser field and produces an attosecond pulse train when the driving pulse has many cycles. The interference of these attosecond pulses results in a frequency comb with an interval of twice the driving field photon energy. When a driving field of a few cycles is used, the cut-off of the spectrum may become a continuum that corresponds to one recollision at the peak of the driving field. A single attosecond pulse was obtained when the continuum portion was extracted [1, 3]. However, for time-resolved absorption spectroscopy and many other applications [4], it is desirable to have a 'white light' source that covers the broad plateau region of the harmonic spectrum. Further more, a single attosecond pulse, instead of a pulse train, is critical for studying the electron dynamics of atoms [5, 6]. To generate the extreme ultraviolet (XUV)

Journal of Modern Optics ISSN 0950–0340 print/ISSN 1362–3044 online
http://www.tandf.co.uk/journals
DOI: 10.1080/09500340410001729573

supercontinuum and single attosecond pulses in the plateau region, the driving pulse duration must be less than one optical cycle, which is very difficult to produce. To avoid such a technical difficulty, the polarization gating technique was proposed [7, 8].

2. The principle of polarization gating

With the three-step model, it is easy to understand that the HHG efficiency decreases rapidly with increasing ellipticity of the driving laser field. The polarization gating takes advantage of this fact. As shown in figure 1, a pulse with a time-dependent ellipticity can be constructed by a left circularly polarized pulse and a delayed right circularly polarized pulse [9]. In the composed pulse, the polarization varies from circular to linear and then back to circular. When such a pulse is used, the oscillating electron will be driven away from the parent ion at both the head and the tail parts by the transverse component of the field. This will eliminate the possibility of re-collision and recombination of the electron with the parent ion [10, 11]. Thus HHG can only be produced by the centre portion of the laser pulse that is nearly linearly polarized.

Assuming that the two input pulses have Gaussian shape with pulse durations τ and the second pulse is delayed by Δt, the time-dependent ellipticity of the combined field in figure 1 is

$$\xi(t) = \frac{\left|1 - e^{-4\ln(2)(t\,\Delta t/\tau^2)}\right|}{1 + e^{-4\ln(2)(t\,\Delta t/\tau^2)}}. \tag{1}$$

According to previous study, the harmonic signal intensity dropped one order of magnitude when the driving field ellipticity changes from 0 to 0.2 [12]. For the following analysis, we assume that only the centre portion of the pulse with $\xi < 0.2$ contributes to the harmonic generation. As shown in figure 1, we call this part the

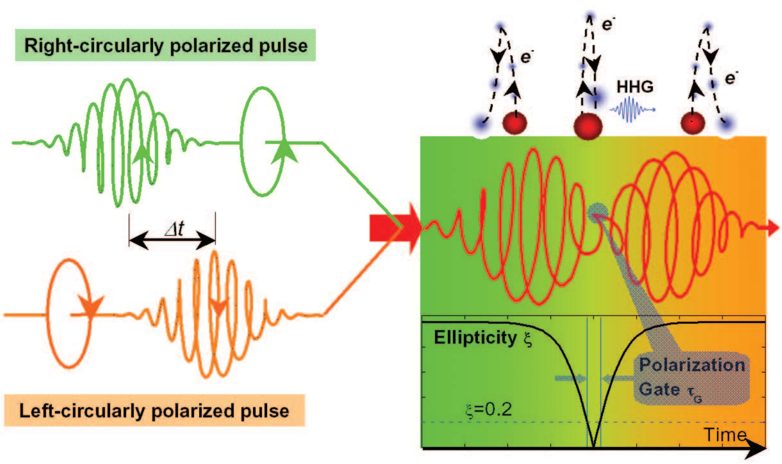

Figure 1. A pulse with a time-dependent ellipticity was formed by a left-circularly polarized pulse and a delayed right-circularly polarized pulse. Under this field, the re-collision process was only possible in the small region at the centre where the ellipticity was close to zero. This region was called the polarization gate.

polarization gate. It gives a measure of the effective pulse length for producing HHG from the pulse with a time-dependent ellipticity. The gate width can be calculated as [13]

$$\tau_G \approx 0.3\frac{\tau^2}{\Delta t}. \tag{2}$$

This gate can be made shorter than one optical cycle by using either very short input pulses or a longer delay time. The former is limited by the shortest laser pulse that one can generate; the latter approach is constrained by the achievable field strength at the linear portion, which can be calculated as

$$E_{\xi=0} = 2E_0 e^{-\{\ln(2)/2\}(\Delta t/\tau)^2}, \tag{3}$$

where E_0 is the peak amplitude of the input circular field. If the delay time is significantly longer than the input pulse duration, the field strength in the gate is much weaker than the input. Thus, the circular field in front of the gate may deplete the ground-state population of the atoms, which suppresses the harmonic generation in the gate. In our experiments, efforts were made to make the laser pulse duration as short as possible so that a short delay could be used.

3. The generation of attosecond extreme ultraviolet supercontinuum

In our experiment, the pulse with a time-dependent ellipticity was constructed by the method illustrated in figure 2 (*a*). The linearly polarized input pulse was evenly divided by a quartz plate into an ordinary ray and an extraordinary ray by setting the optic axis of the quartz plate 45° with respect to the input polarization. Therefore, two pulses with orthogonal polarizations were formed. Because of the birefringence property of the quartz, the ordinary pulse and extraordinary pulse travelled at different group velocities v_o and v_e in the plate. After passing through the plate, the ordinary pulse and the extraordinary pulse were separated in time. The delay was proportional to the plate thickness L. The pulse durations of the two pulses were almost the same because the difference in the group velocity dispersions is small for the two polarization orientations. After the quartz plate, a quarter waveplate was placed with its optic axis along the initial input polarization (45° to the ordinary pulse and the extraordinary pulse). This plate converted the ordinary pulse and extraordinary pulse into left- and right-circularly polarized light respectively. The superposition of these two pulses produced a pulse whose ellipticity changes with time. The method is simple, robust and almost energy lossless. It avoided the difficulties of spatial and temporal overlap of an interferometer-type delay line. Furthermore, there was no additional delay drift induced by the mechanical vibration and the air turbulence of the interferometer.

Figure 2 (*b*) shows the experimental set-up for HHG using the elliptically varying pulse. Part of the output beam from the Kansas Light Source laser system was focused into a hollow-core fibre with a 1 m lens. The fused silica fibre has an inner diameter of 0.25 mm and a length of 1.6 m. It was filled with argon gas with a pressure gradient from about 10^{-1} Torr at the laser entry to about 1 atm at the exit. The collimated output beam from the fibre was reduced to a beam size of about 8 mm to block the ring structure of the high-order modes. The pulse passed through two pairs of chirp mirrors and became negatively chirped with a centre wavelength at 750 nm. This chirp was compensated by a fused silica compensating

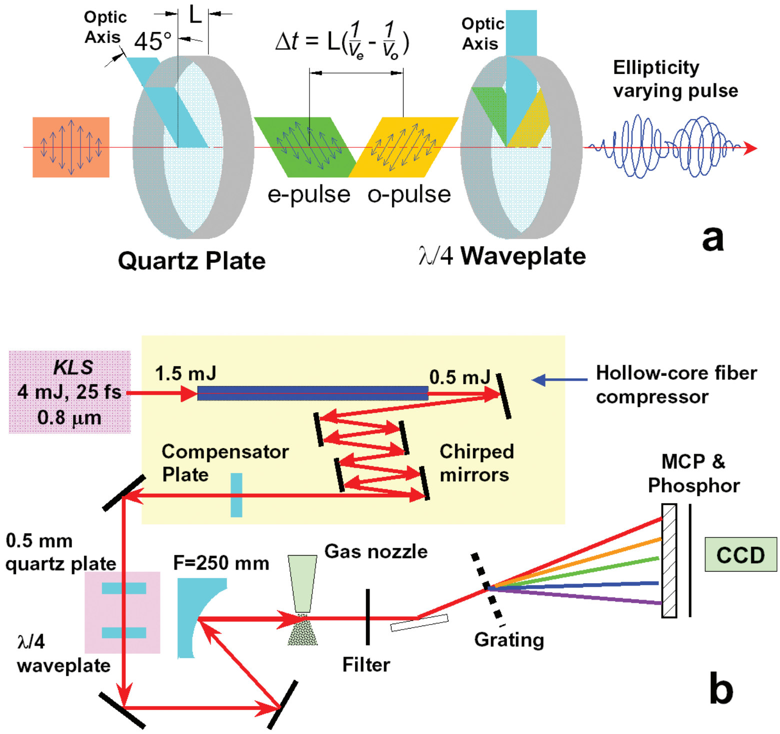

Figure 2. Production of the pulse with a time-dependent ellipticity and its application to the generation of an XUV supercontinuum in the HHG experiment. (*a*) The principle for producing a pulse with a time-dependent ellipticity. (*b*) The overall system layout for HHG: MCP, microchannel plate; CCD, charge-coupled device.

plate so that the pulse duration became tuneable by adjusting the plate thickness. The pulse with a time-dependent ellipticity was then produced with a delay of 15 fs induced by a 0.5 mm quartz plate. Finally the pulse was focused by a parabolic mirror ($f = 250$ mm) into an argon gas jet with a local density of about $10^{18}\,\mathrm{cm}^{-3}$ [14]. The pulse energy was about 260 μJ, which yielded an intensity of about $1.5 \times 10^{14}\,\mathrm{W\,cm}^{-2}$ on the target for the linear portion of the pulse. The intensity is a factor of more than two lower than the saturation ionization intensity of the argon atom in a linearly polarized pulse of about 10 fs duration. The somewhat low intensity is chosen to reduce the ionization of argon atoms by the field before it reaches the linearly polarized portion. The generated HHG signal was dispersed by a transmission grating ($2000\,\mathrm{mm}^{-1}$) and recorded by a microchannel plate detector (MCP) and a cooling charge-coupled device camera.

The harmonic spectra have been measured with different input pulse durations while keeping the delay Δt fixed at 15 fs. According to equation (2) the polarization gate time is proportional to the square of the pulse duration. Figure 3 (*a*) shows the measured pulse duration with a frequency-resolved optical gating set-up as a function of the compensating plate thickness. The pulse duration could be tuned from 8 to 15 fs; this led to the shortest polarization gate width of 1.3 fs, which is about one half-cycle of the driving field. In this case we expected that only one re-collision would occur so that a single attosecond pulse would be produced in the plateau region of the spectrum.

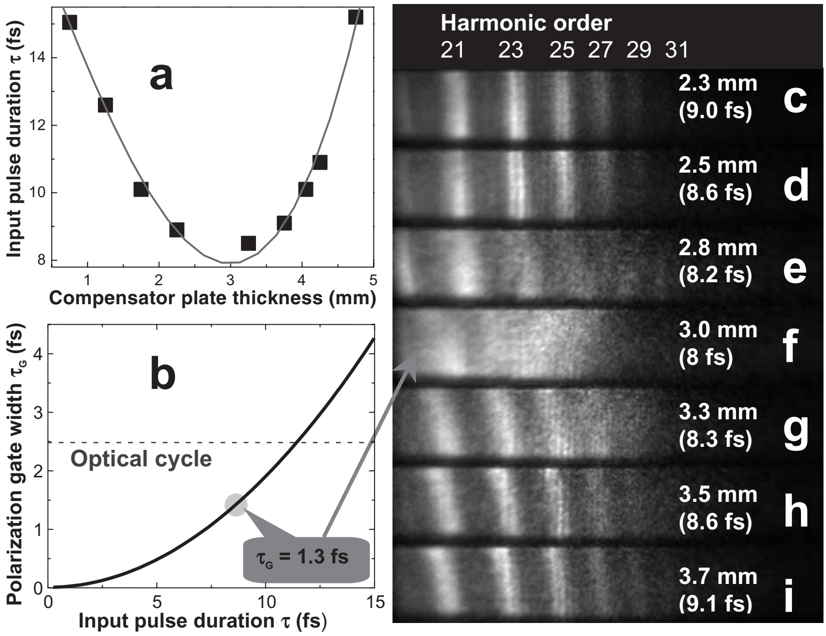

Figure 3. The measured XUV supercontinuum at the HHG plateau and cut-off. (*a*) The measured input pulse duration versus compensating plate thickness. (*b*) The calculated polarization gate width versus input pulse length with an input pulse separation of 15 fs caused by a 0.5 mm quartz plate. (*c*)–(*i*) Measured HHG spectra with compensating plates of different thicknesses: (*c*) 2.3 mm; (*d*) 2.5 mm; (*e*) 2.8 mm; (*f*) 3.0 mm; (*g*) 3.3 mm; (*h*) 3.5 mm; (*i*) 3.7 mm. The corresponding input pulse durations are labelled in the figure.

Figures 3 (*c*), (*d*), (*e*), (*f*), (*g*), (*h*) and (*i*) show the measured HHG spectra taken with the compensating plate thickness of 2.3, 2.5, 2.8, 3.0, 3.3, 3.5 and 3.7 mm respectively. In figure 3 (*c*) each of the circularly polarized pulses for forming the pulse with a time-dependent ellipticity was about 9 fs long. The polarization gate was relatively wide (about 1.6 fs); the HHG spectrum peaks were still resolved. By decreasing the input pulse duration with increasing plate thickness, the HHG peaks gradually merged. In figure 3 (*f*) the thickness of the fused silica fully compensated the frequency chirp, resulting in an 8 fs input pulse and a 1.3 fs polarization gate. The HHG spectrum showed a supercontinuum from 25 to 40 nm, corresponding to harmonic orders from 19 to 31. This result indicates that a single attosecond pulse was generated over the plateau region. On further increasing the compensator plate thickness, the input pulses became longer again. Accordingly, the polarization gate became wider so that the discrete peaks in the HHG spectrum gradually reappeared, which implies that multiple re-collision processes happened.

The line-out of the spectrum in figure 3 (*c*) is shown in figure 4 (*a*). Its Fourier transforms assuming flat phases are shown in figure 4 (*b*). The discrete spectrum resulted in three attosecond pulses with a time interval of a half-cycle of the driving field. The supercontinuum in figures 3 (*f*) and 4 (*c*) yielded a single attosecond pulse with a transform-limited pulse duration of 190 as, as shown in figure 4 (*d*).

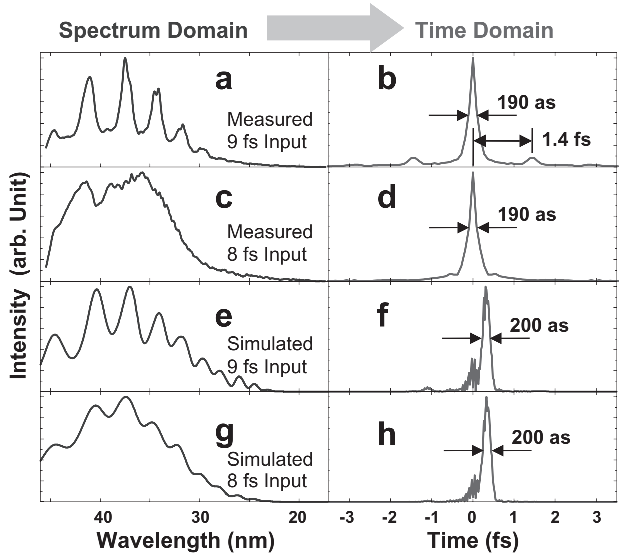

Figure 4. Comparison of experimental results with the numerical simulations in the spectrum domain (left-hand column) and in the time domain (right-hand column). (*a*), (*b*) The line-out of figure 3 (*c*) and its Fourier transform. (*c*), (*d*) The line-out of figure 3 (*f*) and its Fourier transform. (*e*), (*f*) The simulated harmonic spectrum and its Fourier transform with an input pulse duration of 9 fs. (*g*), (*h*) The simulated harmonic spectrum and its Fourier transform with an 8 fs input pulse.

4. Numerical simulation of the polarization gating

Simulations have been made for conditions similar to the experiments. The experimentally measured harmonic signal was a coherent superposition of the dipole radiation from all the atoms in the interaction region. In the numerical simulations, the circularly polarized laser pulses were assumed to be Gaussian in time and in space. The $1/e^2$ spot size at the focus was 30 µm. The harmonic generation from a single atom was calculated using the Lewenstein *et al.* [15] model. The dipole moments in a spatial grid were calculated. The numbers of points were 10 for the propagation direction z and 300 in the radial direction. The number of time steps was 2^{13}. The contributions from all the atoms were summed to obtain the macroscopic harmonic signal using the integral approach developed in [16]. Phase mismatch caused by plasma dispersion was ignored. Our results should be valid when the gas density is very low and the laser intensity is much lower than the saturation ionization intensity.

Figures 4(*e*)–(*h*) illustrate the calculated harmonic spectra and corresponding time domain signals with input pulse durations of 9 fs and 8 fs. The simulations show results similar to the experimental data; that is, the harmonic peaks were resolvable with the 9 fs input and became a supercontinuum when the gate time was close to half the laser period. The time domain results confirmed that a single attosecond pulse was produced with the 8 fs input pulse.

5. Conclusion

We demonstrated that by using a laser pulse of a few cycles from the hollow-core fibre, the linear portion of the ellipticially varying pulse was only half an optical cycle. When this pulse was applied to the HHG experiment, an XUV supercontinuum was generated that corresponded to a single attosecond pulse in the plateau and cut-off spectrum region. To the best of our knowledge, this is the first time that a supercontinuum was obtained using a polarization gating. The ultrafast pulse with a time-dependent ellipticity can be used for other research that involves the re-collision process, such as above threshold ionization of atoms [17] and the double ionization of molecules [18].

Acknowledgments

This work is supported by the Division of Chemical Sciences, Office of Basic Energy Sciences, US Department of Energy. The authors acknowledge Dr Chun Wang and Eric Moon for assistance. We also thank Dr Philip Heimann at Lawrence Berkeley National Laboratory for loaning us the X-ray spectrometer. The Kansas Light Source was built partially with the support of a Major Research Instrumentation (MRI) grant from National Science Foundation awarded to Dr C.L. Cocke and Dr B. DePaola.

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