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Generation of continuum high-order harmonics from carbon plasma using double optical gating

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Abstract
We demonstrated continuum high-order harmonics from carbon plasma using the double optical gating method. The extreme ultraviolet continuum covered 17–25 eV. The observation of such continuum is the first step towards the generation of high-flux single attosecond pulses from plasma harmonics.

(Some figures may appear in colour only in the online journal)

1. Introduction

High-order harmonic generation (HHG) provides a unique source of coherent extreme ultraviolet (XUV) pulses with a wide variety of applications. Examples would include time-resolved photoelectron spectroscopy of organic molecules and solids [1], atomic and molecular spectroscopy [2, 3], interferometry and holography [4] and XUV nonlinear optics [5, 6]. Furthermore, theoretical [7] and experimental works have shown that HHG could be used to produce a train of attosecond pulses [8, 9] or even a single, isolated attosecond pulse [10, 11]. These ultrashort flashes of light have attracted strong interest, since they could record frozen snapshots of ultrafast electron dynamics. Consequently, they are of great interest to a variety of scientific areas, and have already been applied to study photoionization of atoms and molecules with attosecond resolution [12, 13], which has also revealed unexpected features [14, 15]. The generation of an intense, isolated attosecond pulse is of importance for attosecond nonlinear, attosecond pump and attosecond probe experiments. Furthermore, many other applications remain unexplored because of the difficulty to handle and manipulate the harmonics, due partially to the quality of the XUV optics and also due to the relatively low number of photons within an attosecond pulse.

Instead of rare gas atoms, different nonlinear media have been tested in order to increase the conversion efficiency. Due to their high ionization potentials, ions are interesting candidates since they can withstand higher laser intensity and thus can potentially generate both more intense harmonics with higher orders. Such ions can be produced through ionization of rare gases: HHG in Ar^+ was shown to extend the harmonic cut-off to 250 eV [16]. Another candidate for ions is to produce weakly ionized ablation plasma by focusing a relatively long sub-nanosecond laser pulse on a solid target. The harmonics of a time-delayed short laser pulse are then mainly generated from ions within the under-dense plasma [17]. Note that this method is different from that using over-dense plasma, which is also able to produce high harmonics, but through the collective oscillations of the free electrons; this process has been shown recently to result in a train of attosecond pulses [18].

Recently, HHG from low-density plasma has regained attention, following the demonstration of highly efficient HHG from indium plasma (with conversion efficiency of 10^{-4}) [17, 19–23], exceeding conversion efficiencies typically obtained with atomic gases by one order of magnitude [17, 21, 24]. Since then, HHG from more than 15 different target materials has been demonstrated with typical plateau and cut-off features [25, 26]; the highest harmonic order being the 101st from manganese plasma [27]. Different properties of these plasma harmonics have been investigated: the influence

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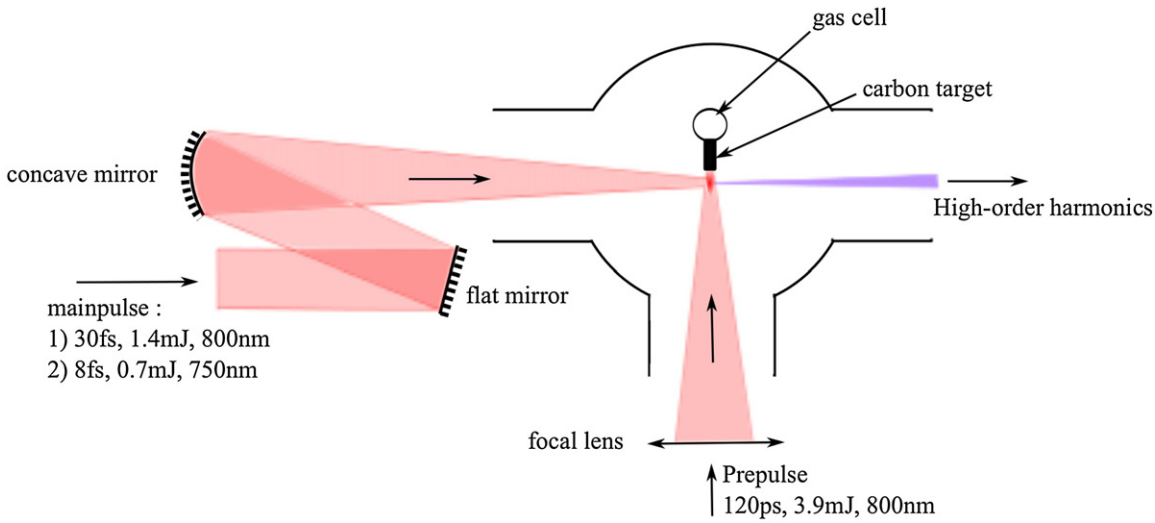

Figure 1. Schematic diagram of the experimental setup for high harmonics generation from under-dense ablation plasma.

of the plasma conditions on the XUV intensity has been studied extensively [17, 28], concluding essentially that a compromise has to be established between the density of singly charged ions, which are mainly responsible for the XUV emission, and that of the free electrons, which degrades phase matching conditions. Elliptical polarization of the driving laser has been shown to completely suppress the XUV emission [22], suggesting the 'usual' three-step process [29] as the origin of the harmonics.

In this paper, we apply the double optical gating (DOG) method [11, 30–32] to the plasma HHG technique, with the goal of demonstrating intense and isolated attosecond pulses. The DOG method is a combination of polarization gating (PG) and two-colour gating. By adding the second harmonic to the PG field, DOG can avoid depletion of the ground-state population of the atom by the leading edge of the laser pulse and make it possible to use driving laser pulses that are two times longer than that in the case of PG alone. The DOG method has been successfully demonstrated to generate isolated attosecond pulses in noble gases [11]. However, the harmonics generated from the noble gas have low conversion efficiency. To take advantage of the demonstrated high conversion efficiency of the high-order harmonics from plasma, and especially from graphitic carbon plasma [34, 35], we propose to couple plasma harmonic generation with the DOG method, to explore the generation of intense isolated attosecond pulses. In this paper, we demonstrate continuum high-order harmonics spectra from ablated carbon plasma. Considering the high conversion efficiency of high-order harmonics from carbon plasma, this would be the first step towards the generation of intense isolated attosecond pulses.

2. Experimental setup

Experiments were performed using the MARS laser [32, 33] at the Kansas Light Source Laboratory of Kansas State University. After compression, the MARS laser delivers laser pulses with 30 fs duration at a repetition rate of 1 kHz and 4 mJ of pulse energy. A schematic diagram of the experimental setup is shown in figure 1. To generate high-order harmonics from low-density plasma, one needs two laser pulses. First, a sub-nanosecond, relatively a low-intensity laser pulse, is used to generate the preformed plasma from a solid target, which will serve as a nonlinear medium for harmonics generation. The initially hot plasma is allowed to expand and cool for some time (typically between 20 and 100 ns). Then, an intense femtosecond driving laser pulse is focused on to the preformed plasma to generate the high-order harmonics. To prepare these two laser pulses, we modified the original configuration of the MARS experiment by splitting the laser beam into two before compression. One beam (120 ps, 3.9 mJ, 800 nm) is used to generate the preformed plasma, while the second beam is sent to a compressor to generate the driving pulse for harmonic generation. For the driving pulse, we had three different configurations. First, we used the compressed pulse as is (30 fs, 1.4 mJ, 800 nm), or second, we further compressed this pulse to 8 fs by passing through a hollow-core fibre and compensating for dispersion using a set of chirp mirrors, resulting in a final pulse energy of 0.7 mJ at a central wavelength of 750 nm. The third configuration is the DOG setup, which is a combination of the PG method with the second harmonic generation of the driving pulse, avoiding depletion of the ground state. This method allows us to use a longer driving pulse, thus opening the possibility to increase the intensity of single attosecond pulses. Experimentally, we used two quartz plates and a BBO crystal to achieve this method.

To directly compare high-order harmonics generated from gas and solid media, we placed the gas cell and the solid carbon target in the same vacuum chamber at the same time. The two targets could be easily switched from one to another without breaking the vacuum, using a vacuum translation stage. To measure the harmonic energy, we used a silicon photodiode (AXUV100, International Radiation Detectors). Several thin aluminium foils were used with the photodiode to eliminate the intense infrared driving laser. To measure the harmonic spectrum, the high-order harmonics are spectrally dispersed using an XUV spectrometer (equipped with a Jobin-Yvon 384 lines mm^{-1} flat-field XUV grating) [36], and a CCD

camera coupled to a computer is used to acquire the harmonic spectra.

Compared with gas harmonic experiments, a major complication with plasma harmonic experiments is the rapid ablation of the solid target material. This causes the target to deform, thus changing the condition of the plasma where the femtosecond driving pulse irradiates. The debris from the ablation also coats the window from which the sub-nanosecond prepulse enters the vacuum chamber, thus reducing the prepulse intensity on target with time. Both of these effects tend to reduce the harmonic intensity. In past experiments with solid metallic targets and using 10 Hz repetition rate lasers, we could generate stable high-order harmonics for 3–5 min, or between 1800 and 3000 shots. However, since current experiments are using kHz repetition rate lasers, this amounts to only several seconds before the harmonic intensity drops, thus making alignment and data acquisition difficult. To avoid such difficulty and to obtain reliable data, we reduced the number of sub-nanosecond prepulses per second on target from 1000 to 20, using an optical shutter (Newport, electronic fast shutter). The optical shutter operates at 10 Hz, but the fastest shutter speed still allowed two pulses from the kHz pulse train to pass. Therefore, data acquisition was performed at an irregular 20 shots s^{-1} repetition rate, comprising two prepulses separated by 1 ms at 10 Hz. The lower prepulse rate allowed us to manually translate the target, thus resulting in stable harmonic generation and longer data acquisition. On the other hand, the 1 kHz femtosecond driving laser was operated without a shutter, since the laser irradiated a position about 100–200 μm from the target surface, ablating very little of the target.

3. Experimental results

3.1. Energy measurement of high-order harmonics

The energy of plasma harmonics using various target material has been measured experimentally [22, 23, 35, 37], showing conversion efficiencies that are higher than those typically observed with gas harmonics. However, there have never been back-to-back measurements that compare the efficiencies of gas and plasma harmonics, using the same pump laser and under the same conditions. Such comparison is important, since the harmonic emission is extremely sensitive to various experimental conditions, including the prepulse intensity and the distance of the driving laser from the target surface. Therefore, we first directly compared the efficiency and characteristics of high-order harmonics from carbon plasma with those from argon gas cell, using the 30 fs laser as the main driving pulse.

Since the quantum efficiency of the photodiode is a function of the wavelength, the first step in evaluating the energy of the high-order harmonics is to calibrate the distribution of the harmonic energy in each harmonic order. Once this information is evaluated for both carbon and argon harmonics, we can calculate the energy in each harmonic order, and the total energy of the high-order harmonics can be evaluated by summing them up.

Table 1. Calculated coefficients for harmonics generated in argon and carbon.

			α_q (%)	
Q	λ_q (nm)	Q_q (%)	Argon	Carbon
11	72.7	1.90	1.24	13.3
13	61.5	2.91	6.18	24.3
15	53.3	3.70	16.8	26.5
17	47.1	4.94	24.2	21.3
19	42.1	5.90	25.8	11.2
21	38.1	6.87	14.2	3.32
23	34.8	7.72	7.37	–
25	32.0	9.49	4.19	–

To evaluate the distribution of the energy among the various harmonic orders, we took the next procedures. First, from the experimentally obtained XUV spectrum, we spatially integrated the harmonic signal within a given wavelength range (around the qth harmonic) for each harmonic order. This is repeated for all orders observed, and the background is subtracted. Next, we calculated the percentage of the harmonic energy that falls into each harmonic order, using the next method: (i) we calculate the area A_v under the voltage curve $V(t)$ versus time obtained using the photodiode; (ii) the energy E_q within the qth harmonic is given by

$$E_q = \frac{A_v \cdot P_q}{R \cdot Q_q}. \tag{1}$$

Here, P_q is the photon energy of the qth harmonic with a wavelength of λ_q, R is the input impedance of the oscilloscope (50 Ω) and Q_q is the quantum efficiency of the photodiode at the wavelength λ_q. Therefore, the total harmonic energy E_{pulse} detected by the photodiode, taking into account the energy distribution in the various harmonics, becomes

$$E_{\text{pulse}} = \sum \alpha_q \frac{A_v \cdot P_q}{R \cdot Q_q}. \tag{2}$$

Here, α_q is the coefficient corresponding to the proportion of the energy in the qth harmonic order. We show in table 1 the values of α_q that were calculated from the spectra.

Using this method, we evaluate the harmonic energy per pulse generated from argon gas and from carbon plasma to be 0.262 and 2.49 nJ, respectively. Therefore, the harmonics generated from carbon plasma have an energy that is 9.5 times higher than those from argon gas cell. Despite this relatively higher energy of the harmonics from carbon plasma, the conversion efficiency seems to be rather low (1.8×10^{-6}), compared with the measured (10^{-4}) values that were recently reported [35]. We attribute this lower efficiency in the current work to the limited energy available in the sub-nanosecond prepulse laser (<4 mJ), which is used to generate the preformed plasma.

Typical plasma harmonic experiments require prepulse laser energy of about 6 to 8 mJ for maximum harmonic output. Since the condition of the preformed plasma (such as the electron and ion density, mean ionization level and plasma dimension) affects greatly the HHG process, it is critical to have the correct pump laser condition as well as the prepulse-to-main-pulse delay time for efficient harmonic generation. In the current experiment, we can conclude that the lower

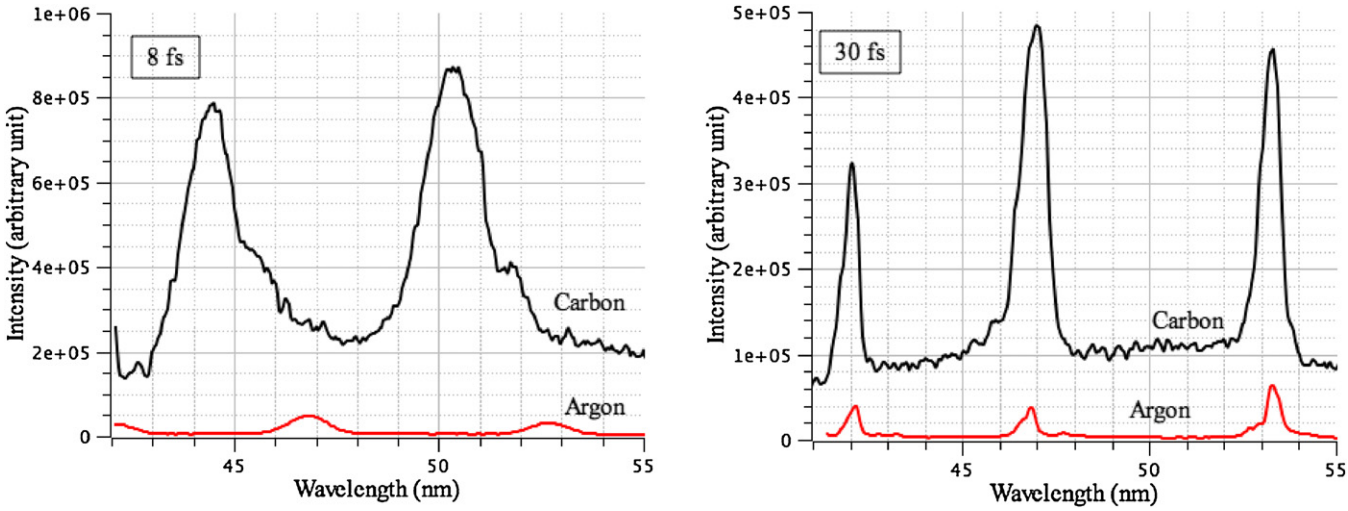

Figure 2. Left: harmonic spectrum generated from carbon plasma (red line) and argon gas (black line) with a 30 fs pulse. Right: harmonic spectrum generated from carbon plasma (red line) and argon gas (black line) with an 8 fs pulse.

prepulse energy did not produce preformed plasma that is optimum for efficient HHG. This conclusion is supported by the fact that we had to focus the femtosecond main pulse laser closer to the target surface than for typical conditions (about 100 μm, compared with the typical 150–250 μm). This indicates that the prepulse was too weak to produce an initial plasma with high enough density, and thus we had to align the driving laser near the target surface. Further, there could also have been strong absorption of the XUV harmonics by the two aluminium filters used in the experiment, which were placed in the XUV beam path to block the intense Ti:sapphire laser beam. The aluminium filters should have a considerable thickness of oxide layer on its surface, which would strongly absorb and attenuate the XUV radiation. Unfortunately, we could not measure the thickness of the oxide layer, and thus no calculation could be done to quantify the actual transmission of the XUV through the filter.

3.2. High-order harmonic spectra from carbon and argon for different pulse duration

Next, we compared the high-order harmonic spectra from argon gas and carbon plasma for the main driving lasers with two different pulse durations, one at 30 fs (1.4 mJ) and the other at 8 fs (0.7 mJ). As explained in section 2, plasma harmonic experiments were performed at an irregular 20 shots s^{-1} acquisition. At this rate, harmonics from carbon plasma was stable enough, thus allowing one to manually translate the target and take reliable data. On the other hand, argon harmonics were acquired at the 1 kHz repetition rate. To compare the relative spectral intensities of the harmonics per shot, the measured harmonic spectra were calibrated for this difference in the repetition rate, as well as to take into account the difference in the acquisition time.

In figure 2, we show the harmonic spectra from carbon plasma and argon gas, whose spectral intensities are adjusted for these differences. For the current experimental setup, harmonics lower than the 15th order are not observed, since the experimental configuration cut these lower order harmonics. We can clearly see from this figure that the harmonic intensities from carbon plasma are much higher than those from the argon gas. The ratio between the harmonic intensities from the two targets is in good agreement with the energy measurements using the silicon photodiodes described in section 3.1.

Another interesting observation is the significant blueshift of the harmonics generated from carbon plasma short driving lasers. For experiments using the 30 fs laser, the high-order harmonics from carbon and argon have the same central wavelength. However, one could observe a significant shift in the harmonics to shorter wavelengths for carbon plasma driven by an 8 fs laser. This phenomenon could be attributed to self-phase modulation of the Ti:sapphire laser within the plasma. Similar effects have been observed in the past, which has been frequently observed when the pump laser irradiates the plasma close (between 50 and 100 μm) to the target surface. The high-density plasma near the target surface could broaden and blueshift the spectrum of the driving laser, which is then reflected in the high-order harmonic spectrum. Since self-phase modulation is a nonlinear effect, its occurrence should be more notable for driving lasers with higher intensities, thus favouring the 8 fs laser.

3.3. Continuum plasma harmonics via double optical gating

Next, we changed the pump laser to a DOG configuration, to demonstrate continuum harmonic generation from plasma created by ablation of a solid carbon target. Figure 3(a) shows harmonics generated using the DOG method, using driving Ti:sapphire lasers with 8 fs pulse duration, 0.7 mJ of energy and 750 nm centre wavelength. This figure clearly shows a continuum harmonic spectrum that differs significantly from those generated by linearly polarized few-cycle pulses (figure 3(b)). The energy of the harmonics in these figures is between 17 and 25 eV. It is difficult to make a direct comparison between these two figures since acquisition parameters were different, but the continuum from carbon plasma was less intense than harmonics generated with the linearly polarized few-cycle pulse.

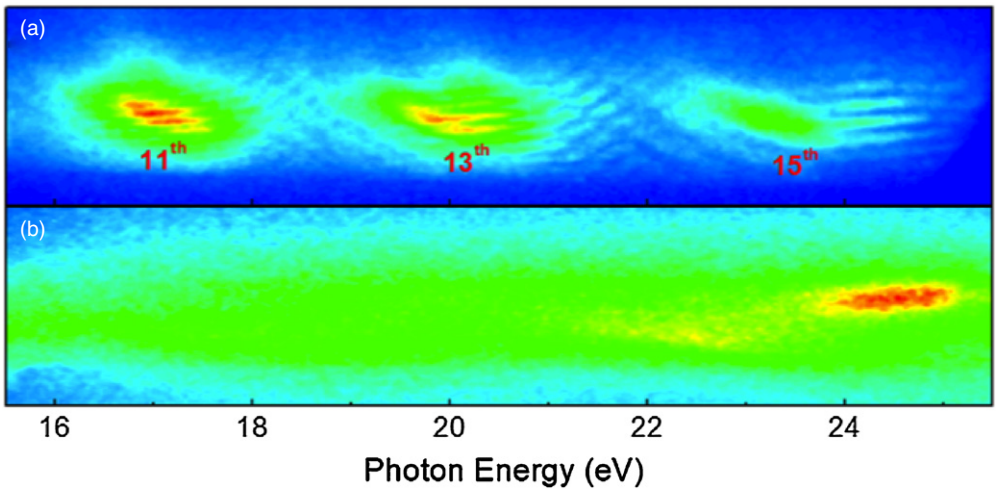

Figure 3. (a) Continuum of harmonics generated in the carbon using DOG method, (b) harmonic generation from carbon using the linearly polarized 8 fs laser pulse only. The colour scales of (a) and (b) are normalized to maximum in each image.

Although a continuum spectrum is not a direct demonstration of the generation of a single attosecond pulse, it is the first step towards the generation of intense isolated attosecond pulses, by combining the plasma harmonic technique and the DOG technique. Currently, the major problem is that we have limited duration for a stable continuum generation due to the manual translation of the solid target surface, and its stability is not long enough for single attosecond pulse characterization or the carrier envelope phase scans. We are currently working on the target design to overcome this problem.

4. Conclusion

Double optical gating was implemented on carbon plasma for the first time. We demonstrated that a continuum of high-order harmonics could be generated with an 8 fs Ti:sapphire laser. Such continuum is a signature of isolated attosecond pulse generation. We are currently modifying our experimental setup to obtain stable harmonics over a larger number of shots, to allow one to verify the generation of isolated attosecond pulses. This promising attosecond pulse generation method opens a large field of nonlinear attosecond applications.

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