Research Article | Vol. 26, No. 25 | 10 Dec 2018 | OPTICS EXPRESS 33238
Optics EXPRESS

Attosecond chirp compensation in water window by plasma dispersion

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Abstract: Compensating attosecond chirp (atto-chirp) of broadband high-order harmonic pulses in the water window region (282 to 533 eV) is a major challenge, due to the lack of natural materials that exhibit negative group velocity dispersion and low loss. Analysis shows that the amount of dispersion of fully ionized hydrogen plasma with suitable density-length product is sufficient to compensate the chirp of attosecond pulses with center photon energy above 300 eV. This is confirmed by numerical simulations based on the Strong Field Approximation.

1. Introduction

High-order harmonic generation in gas media was first discovered near the end of 1980s, which leads to the demonstration of attosecond extreme ultraviolet (XUV) pulses [1,2]. The cutoff photon energy of the single atom high harmonic spectrum can be estimated by

$$\hbar\omega_c = I_p + 3\times10^{-13} I_0 \lambda_0^2, \tag{1}$$

where I_p is the ionization potential of the gas atom, in eV. I_0 and λ_0 are the peak intensity, in W/cm^2, and center wavelength, in μm, of the driving laser [3,4]. It was shown experimentally in 2001 that the cutoff photon energy in the XUV region can be extended by driving high harmonic generation with long wavelength femtosecond lasers [5]. Phase-matched high harmonic generation with mid-infrared lasers was first demonstrated in 2012 [6]. Recently, high flux high harmonic spectra in the water window (282 to 533 eV) have been achieved by using lasers with 1.6 to 2 μm center wavelengths [7]. For generating 533 eV X-rays, i.e., the Oxygen K-edge, at the cutoff of high harmonics by interacting helium atoms with laser pulses centered at 1.7 μm, the peak intensity of the infrared laser needs to be higher than 6×10^{14} W/cm^2.

It is well known that attosecond pulses from high harmonic generation are chirped. The signs of atto-chirp for short and long trajectories are different. However, only X-ray emissions from short trajectory can be phase matched on the propagation axis [8]. The atto-chirp corresponding to the short trajectory is positive. The value at the center of the plateau region of the high harmonic spectrum may be estimated by Eq. (2) that is derived by using the semi-classical three-step model [9],

$$Chirp = 1.63\times10^{18} \frac{1}{I_0 \lambda_0}. \tag{2}$$

The unit of the chirp is as^2, and the units of I_0 and λ_0 are the same as in (1). The chirp is 1600 as^2 when the 1.7 μm IR driving laser intensity is at 6×10^{14} W/cm^2. The atto-chirp slowly increases towards both ends of the spectrum. The effects of the driving laser wavelength on attosecond chirp have been previously investigated experimentally [10].

#349535 https://doi.org/10.1364/OE.26.033238
Journal © 2018 Received 30 Oct 2018; revised 17 Nov 2018; accepted 17 Nov 2018; published 5 Dec 2018

The positive atto-chirp in the wavelength range below 300 eV can be compensated by the negative group delay dispersion (GDD) of thin foils or neutral gases [11]. Recently, isolated attosecond X-ray pulses around the Carbon K-edge (282 eV) has been characterized [12], where the atto-chirp is reduced by Sn thin films with 400 nm total thickness. It is, however, difficult to find proper materials that have sufficient dispersion above 300 eV. Chirped CrSc multilayer mirrors with GDD = -8000 as^2 in the water window region have been designed, but the bandwidth is narrow (<30 eV) and the peak reflectivity is low (~1%) at 326 eV center photon energy [13]. The detuned zero-dispersion grating stretcher (two gratings and a telescope in the 4f configuration) is another scheme to introduce negative GDD. A single pass stretcher for 25 eV bandwidth XUV pulses at 73 eV has been designed where the spatial chirped is neglected [14]. For broadband X-ray pulses in the water window, it is challenging to achieve high throughput considering the limited diffraction efficiency of gratings in the 282 to 533 eV region and the total reflectivity of the three extra mirrors added to the optical path (one for beam collimation and two for the telescope). With the aim of compensating atto-chirp in the water window with low loss, we analyzed the GDD of plasma for soft X-rays.

2. Dispersion of plasma in the soft X-ray region

The index of refraction of fully ionized hydrogen plasma in the soft X-ray region is

$$n(\omega_x) = \sqrt{1 - \frac{\omega_p^2}{\omega_x^2}}, \tag{3}$$

where ω_x and ω_p are the angular frequencies of the X-ray and plasma respectively. The latter in determined by the number density of electrons N_e,

$$\omega_p = \sqrt{\frac{e^2 N_e}{\varepsilon_0 m_e}}, \tag{4}$$

where e and m_e are the charge and mass of the electron respectively. ε_0 is the permittivity of free space. Equations (3) and (4) are also valid for free protons except that the mass in (4) needs to be replaced by that of a proton. They indicate that the contribution to the index of refraction by the protons in the plasmas is three orders of magnitude smaller than that of electrons.

For the plasma density considered here, $\omega_x >> \omega_p$,

$$n(\omega_x) \approx 1 - \frac{1}{2}\frac{\omega_p^2}{\omega_x^2}. \tag{5}$$

The group velocity of plasma can be derived from the dispersion relation $\omega_x^2 = k_x^2 c^2 + \omega_p^2$,

$$v_g(\omega_x) = \frac{d\omega_k}{dk_x} = n(\omega_x)c, \tag{6}$$

where k_x is the propagation constant and c is the speed of light in vacuum. The group velocity dispersion of the plasma is

$$GVD(\omega_x) \approx -\frac{\omega_p^2}{c\omega_x^3} = -\frac{e^2}{\varepsilon_0 m_e c}\frac{N_e}{\omega_x^3}, \tag{7}$$

which is negative. The group delay dispersion (GDD) of a plasma column with length L_p is

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$$GDD(\hbar\omega_x) = -\frac{e^2\hbar^3}{\varepsilon_0 m_e c}\frac{N_e L_p}{(\hbar\omega_x)^3}. \tag{8}$$

Apparently it is the electron density-length product that determines the GDD value.

The particle number density of a standard gas at 1 atmosphere pressure and room temperature is 2.5×10^{19} /cm^3. When H_2 molecules under such conditions are fully ionized, $N_e = 5\times10^{19}$ /cm^3. The GDD of such plasma with $L_p = 10$ cm is -559 as^2 at $\hbar\omega_x = 300$ eV, which can be used to compensate the atto-chirp of the water window X-ray pulses generated by a 1.7 μm laser. The length of the plasma for compensating a 1600 as^2 chirp at 365 eV is about 54 cm. The transmission of fully ionized hydrogen plasma with the required density-length product is almost 100% for X-rays above 300 eV. Hydrogen or helium plasma are preferred to avoid absorption of X-rays by inner shells of atoms.

3. Numerical simulations of chirp compensation

Numerical simulations based on the Strong Field Approximation of high harmonic generation [15] have been performed to demonstrate feasibility of the atto-chirp compensation by the dispersion of plasma. The dipole moment of a single helium atom in the time-domain was calculated using the open-source code, HHGmax [16]. The dipole matrix element is hydrogen-like and the ionization potential is 24.59 eV. The contribution from the long trajectory is suppressed by properly setting the integration and window parameters in the code. The pulsed external laser field has a Gaussian temporal profile, as shown in Fig. 1(a). The FWHM duration, carrier envelope phase (CEP) and peak intensity are 6 fs, 5π/8 rad, and 7×10^{14} W/cm^2 respectively. The pulse duration and CEP values are chosen to demonstrate the generation of single isolated attosecond pulses with the amplitude gating [17].

The power spectrum in the 300 to 500 eV range are shown in Fig. 1 (b). The continuous-like spectrum is the result of the single recollision of the freed electron with the parent ion in the quasi-single cycle laser field. The small fast modulations in the power spectrum are due to beating between the main pulse and a very week pre-pulse one half IR cycle ahead. The quadratic phase of the X-ray directly from the atom (without chirp compensation) reveals a positive chirp, as illustrated in Fig. 1(c). When the phase of a 54 cm long fully ionized plasma column with $N_e = 5\times10^{19}$ /cm^3 electron density is added to the high harmonic phase, the chirp is significantly reduced, which can be seen by the close to flat total phase in Fig. 1(c). The chirp compensation is not perfect and the shape of the total phase curve suggests that the remaining phase errors are dominated by the 3rd order dispersion. The electron density-length product value was chosen to yield the shortest compressed X-ray pulse, which is consistent with the estimates in section 2.

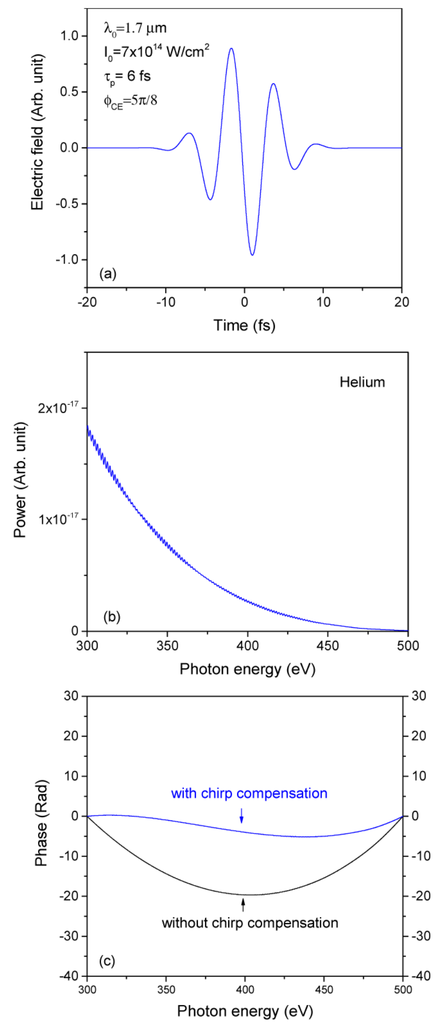

Fig. 1. (a) Driving laser field. (b) Power spectrum of the X-ray pulse. (c) Phases of X-rays with and without chirp compensation by plasma dispersion.

Without applying the chirp compensation, the electric field of the X-ray pulse corresponding to the spectrum above 300 eV is illustrated in Fig. 2 (a), which is far from transform limited. When the plasma phase is added, the X-ray pulse duration is significantly reduced as a result of the atto-chirp compensation by the plasma dispersion, as shown in Fig. 2(b). The compressed X-ray pulse contains only two cycles, which is close to the transformed limited case, Fig. 2(c). The asymmetry of the waveform in Fig. 2(b) is due to the incompleteness of the chirp compensation. The slow decay of the trailing edge is caused by the third order phase error.

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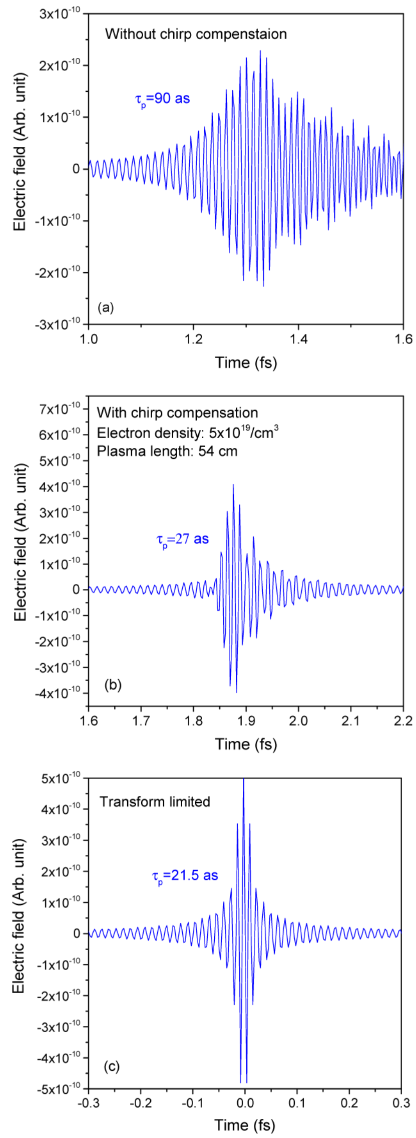

Fig. 2. Electric fields of the X-ray pulses. (a) Without chirp compensation. (b) With chirp compensation. (c) Transform-limited. The pulse durations, τ_p, are the FWHM of the intensity profiles.

It is technically feasible to construct a plasma column that offers the required electron density-length product. Hydrogen plasma waveguides have been developed for accelerating electrons with the laser wakefield [18,19]. The plasma is formed by pulsed high voltage and high current discharges through a gas-filled capillary. The typical length of the plasma column is 1 to 10 cm and the electron number density varies from 10^{17} to 10^{19} cm^3. The range of the diameter of the capillary is 0.1 to 1 mm. The repetition rate is limited by the capacity of the power supply and 1 kHz operation has been demonstrated [20].

The electron density-length product for chirp compensation in the water window is larger than that for electron acceleration, which requires more powerful discharge power supplies [21] or propagating the X-ray beam through multiple stages of plasma columns. Since guiding

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of the intense infrared laser beam is not required for attosecond chirp compensation, other discharge configurations may be considered.

4. Summary

It is shown that the group delay dispersion of fully ionized plasma columns with 10^{19} cm^3 electron density and tens of cm in length is sufficient to compensate attosecond chirp in the water window and produce few-cycle X-ray pulses. The compressed pulse duration approaches one atomic unit of time (24.2 as). It was predicted that when an electron is removed from multi-electron atoms or molecules, it takes about 50-as for other electrons to response [22]. Attosecond water window X-ray pulses achieved with chirp compensation may serve as probes to study such ultrafast electron dynamics in molecules that contains C, N and O atoms. Being negative definite, the plasma dispersion may also be used at other photon energy ranges. Since the GDD is inversely proportional to the cube of the X-ray photon energy, much larger electron density-length product is needed to compensate atto-chirp at the keV range.

Funding.

Air Force Office of Scientific Research (AFOSR) (FA9550-15-1-0037, FA9550-16-1-0013, FA9550-17-1-0499); Army Research Office (ARO) (W911NF-14-1-0383); Defense Advanced Research Projects Agency (DARPA) PULSE (W31P4Q1310017). National Science Foundation (NSF) (1806584).

References

1. P. M. Paul, E. S. Toma, P. Breger, G. Mullot, F. Augé, P. Balcou, H. G. Muller, and P. Agostini, "Observation of a train of attosecond pulses from high harmonic generation," Science **292**(5522), 1689–1692 (2001).
2. M. Hentschel, R. Kienberger, C. Spielmann, G. A. Reider, N. Milosevic, T. Brabec, P. Corkum, U. Heinzmann, M. Drescher, and F. Krausz, "Attosecond metrology," Nature **414**(6863), 509–513 (2001).
3. P. B. Corkum, "Plasma perspective on strong field multiphoton ionization," Phys. Rev. Lett. **71**(13), 1994–1997 (1993).
4. K. J. Schafer, B. Yang, L. F. DiMauro, and K. C. Kulander, "Above threshold ionization beyond the high harmonic cutoff," Phys. Rev. Lett. **70**(11), 1599–1602 (1993).
5. B. Shan and Z. Chang, "Dramatic extension of the high-order harmonic cutoff by using a long-wavelength driving field," Phys. Rev. A **65**(1), 011804 (2001).
6. T. Popmintchev, M.-C. Chen, D. Popmintchev, P. Arpin, S. Brown, S. Alisauskas, G. Andriukaitis, T. Balciunas, O. D. Mucke, A. Pugzlys, A. Baltuska, B. Shim, S. E. Schrauth, A. Gaeta, C. Hernandez-Garcia, L. Plaja, A. Becker, A. Jaron-Becker, M. M. Murnane, and H. C. Kapteyn, "Bright coherent ultrahigh harmonics in the keV x-ray regime from mid-infrared femtosecond lasers," Science **336**, 1287–1291 (2012).
7. X. Ren, J. Li, Y. Yin, K. Zhao, A. Chew, Y. Wang, S. Hu, Y. Cheng, E. Cunningham, Y. Wu, M. Chini, and Z. Chang, "Attosecond light sources in the water window," J. Opt. **20**(2), 023001 (2018).
8. S. Kazamias and P. Balcou, "Intrinsic chirp of attosecond pulses: Single-atom model versus experiment," Phys. Rev. A **69**(6), 063416 (2004).
9. Z. Chang, "Fundamentals of Attosecond Optics," ISBN 9781420089370 (2011).
10. G. Doumy, J. Wheeler, C. Roedig, R. Chirla, P. Agostini, and L. F. DiMauro, "Attosecond synchronization of high-order harmonics from midinfrared drivers," Phys. Rev. Lett. **102**(9), 093002 (2009).
11. K. T. Kim, C. M. Kim, M.-G. Baik, G. Umesh, and C. H. Nam, "Single sub-50- attosecond pulse generation from chirp-compensated harmonic radiation using material dispersion," Phys. Rev. A **69**(5), 051805 (2004).
12. J. Li, X. Ren, Y. Yin, K. Zhao, A. Chew, Y. Cheng, E. Cunningham, Y. Wang, S. Hu, Y. Wu, M. Chini, and Z. Chang, "53-attosecond X-ray pulses reach the carbon K-edge," Nat. Commun. **8**(1), 186 (2017).
13. A. Guggenmos, R. Rauhut, M. Hofstetter, S. Hertrich, B. Nickel, J. Schmidt, E. M. Gullikson, M. Seibald, W. Schnick, and U. Kleineberg, "Aperiodic CrSc multilayer mirrors for attosecond water window pulses," Opt. Express **21**(19), 21728–21740 (2013).
14. L. Poletto, F. Frassetto, and P. Villoresi, "Design of an extreme-ultraviolet attosecond compressor," J. Opt. Soc. Am. B **25**(7), B133–B136 (2008).
15. M. Lewenstein, P. Balcou, M. Y. Ivanov, A. L'Huillier, and P. B. Corkum, "Theory of high-harmonic generation by low-frequency laser fields," Phys. Rev. A **49**(3), 2117–2132 (1994).
16. M. Hoegner, https://github.com/Leberwurscht/HHGmax
17. M. Chini, K. Zhao, and Z. Chang, "The generation, characterization and applications of broadband isolated attosecond pulses," Nat. Photonics **8**(3), 178–186 (2014).

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18. D. J. Spence and S. M. Hooker, “Investigation of a hydrogen plasma waveguide,” Phys. Rev. E Stat. Nonlin. Soft Matter Phys. **63**(1 Pt 2), 015401 (2001).
19. D. Spence, A. Butler, and S. M. Hooker, “Gas-filled capillary discharge waveguides,” J. Opt. Soc. Am. B **20**(1), 138–151 (2003).
20. A. Gonsalves, F. Liu, N. Bobrova, P. Sasorov, C. Pieronek, J. Daniels, S. Antipov, J. Butler, S. Bulanov, W. Waldron, D. E. Mittelberger, and W. P. Leemans, “Demonstration of a high repetition rate capillary discharge waveguide,” J. Appl. Phys. **119**(3), 033302 (2016).
21. A. E. Dyson, C. Thornton, and S. M. Hooker, “A compact, low cost Marx bank for generating capillary discharge plasmas,” Rev. Sci. Instrum. **87**(9), 093302 (2016).
22. J. Breidbach and L. S. Cederbaum, “Universal attosecond response to the removal of an electron,” Phys. Rev. Lett. **94**(3), 033901 (2005).