Self-Compression of Attosecond High-Order Harmonic Pulses

Article *in* Physical Review Letters · December 2007

DOI: 10.1103/PHYSREVLETT.99.223904 · Source: PubMed

6 authors, including:

Kyung Taec Kim
Gwangju Institute of Science and Technology
75 PUBLICATIONS **1,026** CITATIONS
SEE PROFILE

Mi Na Park
41 PUBLICATIONS **240** CITATIONS
SEE PROFILE

Tayyab Imran
COMSATS University Islamabad
46 PUBLICATIONS **188** CITATIONS
SEE PROFILE

Self-Compression of Attosecond High-Order Harmonic Pulses

Kyung Taec Kim,[1] Kyung Sik Kang,[1] Mi Na Park,[1] Tayyab Imran,[1] G. Umesh,[2] and Chang Hee Nam[1]

[1]*Department of Physics and Coherent X-Ray Research Center, Korea Advanced Institute of Science and Technology (KAIST), Daejeon 305-701, Korea*

[2]*Physics Department, National Institute of Technology Karnataka, Surathkal, Mangalore 575025, India*

(Received 28 July 2006; published 29 November 2007)

Self-compression of attosecond high-order harmonic pulses in the harmonic generation medium itself has been demonstrated. The attosecond pulses were generated in an argon-filled gas cell and compressed by exploiting the dispersion characteristics of argon. Since the harmonic generation medium itself was used as the compression medium, continuous chirp control was easily achieved by adjusting the gas pressure. The optimized attosecond pulse was also the most intense, and its duration of 206 as was very close to the transform-limited value of 200 as.

DOI: 10.1103/PhysRevLett.99.223904 PACS numbers: 42.65.Ky, 32.80.Rm

Generation of attosecond pulses using high-order harmonic processes has been extensively investigated in recent years. This has opened up the new areas of research called attosecond physics and attosecond science [1]. Progress in harmonic generation theory and relevant experimental measurements have led to deeper insight into the harmonic generation processes. It is now an established fact that the attosecond harmonic pulses are positively chirped due, mainly, to the quadratic spectral phase variation [2–4]. Compensation of the intrinsic chirp contained in the attosecond pulses has been a topic of active research since it limits the achievement of transform-limited attosecond pulses. Some of the conventional techniques, used to manipulate the chirp of laser pulses at visible wavelengths, may be extended to the extreme ultraviolet region. A chirped multilayer x-ray mirror can be used for chirp compensation [5], but the fabrication of such a mirror is rather difficult. Chirp compensation by using the dispersion property of an x-ray filter material—thin metal film—was theoretically proposed [2,3] and was experimentally implemented recently [6].

In this Letter, we report on the chirp compensation of attosecond harmonic pulses by exploiting the dispersion property of the harmonic generation medium itself. As the chirp compensation is achieved in the harmonic generation medium, no additional component is necessary for chirp control. In our experiments, we obtained an attosecond pulse train with a pulse width, which is very close to the transform-limited width at optimized conditions.

The formation of intrinsic chirp in an attosecond pulse may be well explained by the semiclassical three-step model, which implies that the harmonic photon energy increases with time in the case of short trajectory harmonics and decreases in the case of long trajectory harmonics [4,7,8]. Consequently, harmonics are positively chirped for short trajectory components and negatively chirped for long trajectory components. As the experimental conditions can be easily set to favor the short trajectory process for strong harmonic generation, attosecond harmonic pulses are positively chirped [9,10]. Since the occurrence of the attosecond pulse chirp is due to a sinusoidal laser electric field, it is intrinsic and unavoidable in a single atom calculation.

When the emitted harmonics propagate through a medium, the dispersion characteristics of the medium can alter the intrinsic chirp of the attosecond pulse. In particular, positively chirped attosecond harmonic pulses can be compensated if the propagating medium has negative group delay dispersion (GDD). According to the Kramers-Kronig relations, the dispersion characteristics of a medium, obtained from the refractive index calculations, exhibits negative GDD in the frequency range wherein increasing transmission follows a strong absorption [7]. Some rare gases, such as Ar, Kr, and Xe, commonly used for high-order harmonic generation (HHG), exhibit such transmission and dispersion characteristics. These gases can, therefore, be used for chirp compensation in addition to strong harmonic generation. Figure 1 shows the spectral transmittance and GDD of a 1-mm-long Ar gas column at a pressure of 40 torr. The transmittance is very low below the 26th harmonic order and rapidly increases for higher orders [11]. This shows that the GDD is negative in the wave-

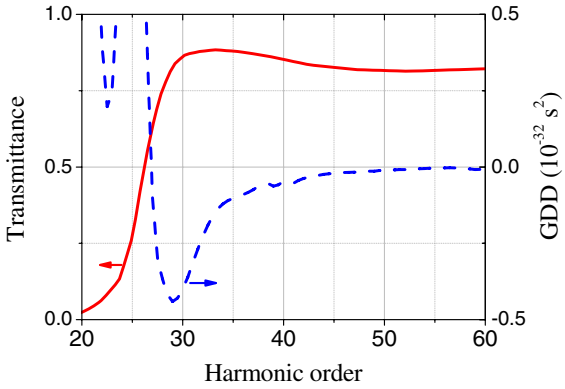

FIG. 1 (color online). Spectral transmittance (solid line) and GDD (dashed line) of the 40-torr 1-mm-long Ar gas column.

length region above the 26th order where, additionally, strong harmonic generation can be achieved [12]. For the lower order harmonics this dispersion property may adversely enhance the positive chirp; however, the harmonics in this range are strongly absorbed. Therefore, when the dispersion characteristics of the medium are properly taken into account, self-compression of attosecond harmonic pulses can occur in the harmonic generation medium itself.

For simultaneous achievement of strong harmonic generation and chirp compensation, a long gas cell was specially designed and used. The gas cell consists of three parts for high pressure operation, as shown in Fig. 2. The first and third cells were subjected to differential pumping to maintain a low gas pressure in the experimental chamber. The length of the middle gas cell was determined to be 12 mm so as to achieve the compensation of the intrinsic chirp for the harmonics higher than the 26th order. The schematic of the experimental setup is shown in Fig. 2. A kHz Ti:sapphire laser, generating pulses of 30-fs duration at 815 nm, was used to generate high-order harmonics. The laser beam was split into two by a beam splitter. Using a concave mirror of f number 60, the first beam was focused into the middle gas cell for generating high-order harmonics. The entrance of the middle cell was located 4 mm behind the focus. The second beam was made to interfere with the harmonics to enable temporal characterization of the harmonics. The laser intensity was about 2×10^{14} W/cm^2 for the harmonic generation and 1×10^{12} W/cm^2 for the probe beam. After harmonic generation, the transmitted laser beam was blocked by a 200-nm aluminum filter to completely eliminate the laser light. The harmonic and the probe beams were combined using a mirror having a hole in the center and both beams were then focused together, using a gold-coated toroidal mirror, into a photoionization gas chamber. The generated photoelectrons were analyzed with a magnetic-bottle time-of-flight electron spectrometer.

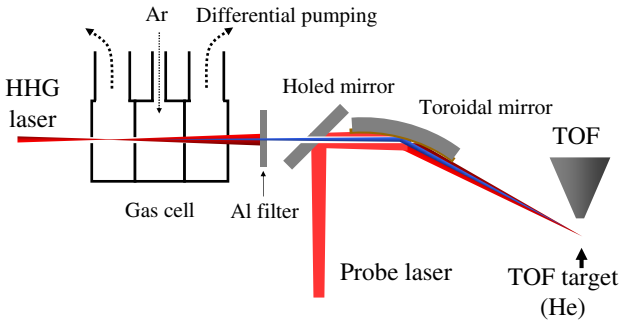

FIG. 2 (color online). Schematic of the experimental setup. The gas cell designed for high pressure operation is also shown. The 45° mirror, which has a 1-mm diameter hole at the center, is used to facilitate overlapping of the harmonic and the probe beam. They are focused again into the target of the time-of-flight spectrometer by a gold-coated toroidal mirror.

The intrinsic chirp of the attosecond pulse can be compensated by the negative GDD of an appropriately chosen medium. The amount of GDD can be controlled by both the medium length and pressure. In our experiments the medium length was set to 12 mm, and gas pressure was adjusted to compensate the intrinsic chirp of the attosecond pulses. In order to observe the chirp variation with gas density, the spectral phase difference between harmonics should be measured. Generated attosecond pulses were characterized by the method of reconstruction of attosecond beating by interference of two photon transitions (RABITT). In this method the two-photon, two-color ionization of the gas atoms by the harmonic and the probe beam pulses is produced. The resultant photoelectron spectrum shows the occurrence of side bands between the direct photoionization signals due to the harmonics. This provides the information on the spectral phase difference between adjacent harmonics. The amplitude of the sideband signal is modulated with varying time delay, τ, between the probe beam and harmonic pulses. By fitting the sideband amplitude modulation to the expression $\cos(2\omega_0\tau + \varphi_{q-1} - \varphi_{q+1})$, one can obtain the spectral phase difference, $\Delta\varphi_q$, between the spectral phase of the $(q-1)$th harmonic φ_{q-1} and that of the $(q+1)$th harmonic φ_{q+1} [13]; here ω_0 is the frequency of the probe laser. The amplitude information of the harmonics can be obtained from the photoelectron spectrum generated without the probe beam by taking into account the known photoionization cross sections. The phase and the amplitude information so obtained allow the reconstruction of the waveform of an attosecond pulse train. As this method assumes that the photoelectron spectrum has a comblike structure, the reconstructed temporal structure represents that of a pulse in an infinite attosecond pulse train. In other words, the reconstructed attosecond pulse really represents only an averaged pulse. However, the spectral phase information obtained from the RABITT analysis is sufficient for analyzing the chirp condition of an attosecond pulse train.

For the RABITT measurements, first the spectral amplitude of harmonics was obtained from the photoelectron spectrum of helium gas without using the probe beam. Since helium gas has a high ionization potential, the lowest harmonic contributing to the photoionization of He would be the 17th harmonic. The photoelectron distribution measured at different argon pressures is presented in Fig. 3 for photoelectrons generated by high-order harmonics from 17th to 41st order. For the 15-torr case, the intensities of the lower harmonics are comparable with those at higher harmonics even though the transmittance is quite low, indicating that the harmonic generation occurred throughout the entire medium. The maximum photoelectron signal was observed at 25th, 27th, and 29th harmonics for argon pressure of 15, 30, and 40 torr, respectively. It is seen that the maximum harmonic intensity shifts toward higher harmonic orders with increasing gas density. This is due to the

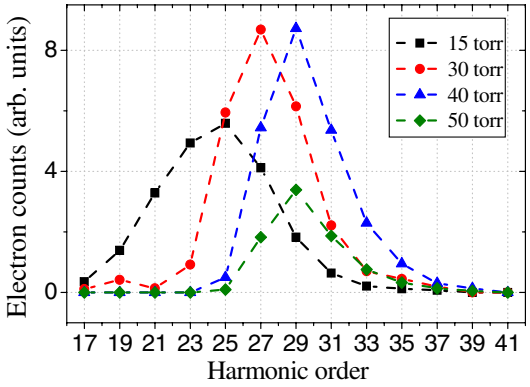

FIG. 3 (color online). Photoelectron counts from He ionized by high-order harmonics from argon at a gas pressure of 15 torr (rectangles), 30 torr (circles), 40 torr (triangles), and 50 torr (diamonds).

fact that the absorption of harmonics in argon decreases with increasing harmonic order in this energy range, as shown in Fig. 1, generating strong harmonics of higher order at higher gas pressures.

For the spectral phase analysis, photoelectron spectra were obtained using harmonics and the probe beam, increasing the probe time delay in steps of about 200 as. The measurements were performed for argon pressure varying from 15 to 50 torr. The 15-torr and 40-torr cases are shown in Fig. 4(a). The sideband modulations, seen clearly in Fig. 4(a), were fitted to a cosine function, and the results were analyzed in terms of the group delay. The group delay, given by the spectral phase difference $\Delta\varphi$ between neighboring harmonics divided by $2\omega_0$, are shown in Fig. 4(b). The offset of the group delay, corresponding to the linear spectral phase, does not affect the pulse shape, and, hence, was adjusted to zero at the 26th sideband for better visualization. The positive slope here implies that the harmonic pulses possess positive chirp. In the 15-torr case, the second-order spectral phase, measured by the slope of the linear fit from the 26th to the 36th order, is about 1.3×10^{-32} s^2. As the gas density increases up to 30 torr, the positive chirp increases for harmonic orders below the 26th due to the positive GDD of Ar as shown in Fig. 1. The behavior of harmonics higher than the 26th order remains similar to that for the 15-torr case, indicating that the single atom feature is still dominant.

As the pressure in the gas cell is increased above 40 torr, the spectral transmittance and dispersion characteristics affect even the higher order harmonics. Harmonics below the 26th order are severely attenuated due to the strong absorption in argon as shown in Fig. 1. As seen in Fig. 4(b), the slope of the group delay changes from negative to positive value around the 31st order, mainly due to the third-order spectral phase induced by the third-order dispersion of Ar. The second-order spectral phase, obtained by averaging the slope of the delay, is only 1.7×10^{-33} s^2 for the 40-torr case. This implies that the intrinsic chirp of

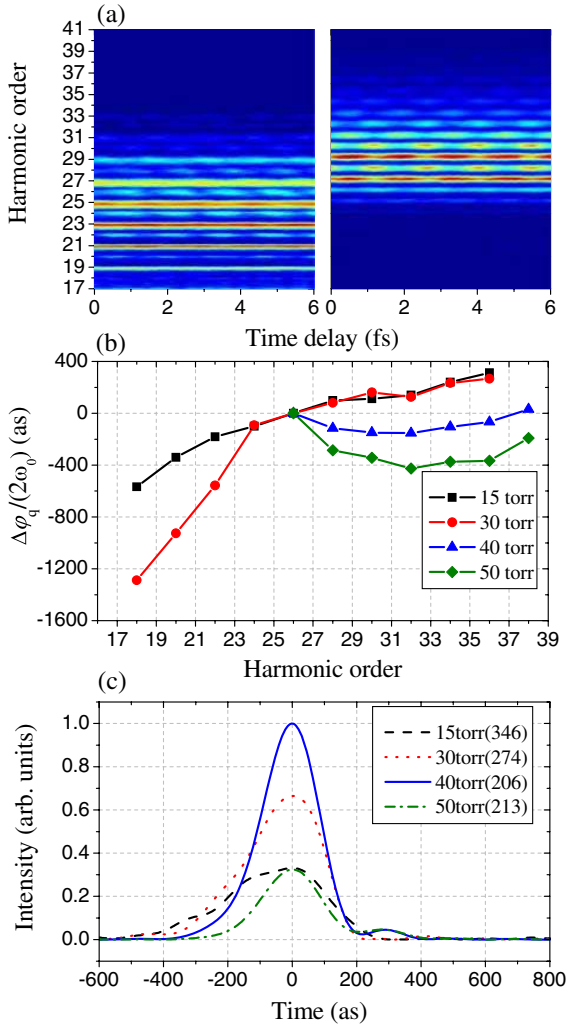

FIG. 4 (color online). (a) Evolution of the photoelectron spectra with time delay in the cases of argon pressure equal to 15 torr (left) and 40 torr (right). (b) Group delay, $\Delta\varphi_q/2\omega_0$, obtained using the RABITT method, for the different gas pressures (same as in Fig. 3). (c) Reconstructed attosecond pulse profiles. The pressure and the pulse widths in FWHM are shown in parentheses in the legend.

the attosecond pulses is well compensated. Since the gas cell was placed beyond the laser focus, the laser beam entering the medium was slightly diverging. The laser interaction with the medium would increase the beam divergence further with increasing gas density because of the plasma defocusing effect in an ionized medium [14]. In such a situation, the harmonic generation would occur, primarily, in the beginning of the gas cell, and the positive chirp of the attosecond pulse gets compensated as the beam propagates further in the gas, on account of the negative GDD of the medium itself. The effective absorption length may be estimated from the change in GDD for the 40-torr case wherein the pulse compression occurs. The GDD of 1-mm-long, 40-torr Ar gas, averaged over the harmonic range from 26th to 38th order, is obtained to be

-0.24×10^{-32} s^2. This indicates that, in the 40-torr case, about a 5-mm portion of the gas cell contributed to the chirp compensation. If a metallic x-ray filter were to be used for the chirp compensation, the required thickness of an aluminum filter, which is most suitable for this wavelength range, would be 2.8 μm. When the gas pressure was increased further to 50 torr, the second-order spectral phase, estimated from Fig. 4(b), is -0.57×10^{-32} s^2. Thus, the intrinsic chirp was overcompensated and the attosecond pulse train became negatively chirped. Further, the weakened photoelectron spectrum indicates that the harmonics were strongly absorbed during propagation.

From the RABITT measurements, temporal profiles of attosecond harmonic pulses were reconstructed, as shown in Fig. 4(c). The pulse widths, i.e., the full width at half maximum (FWHM), are calculated to be 348, 295, 206, and 213 as for the 15-, 30-, 40-, and 50-torr cases, respectively. We obtained the shortest harmonic pulse of 206 as in the 40-torr case. As its transform-limited width, estimated from the harmonic signal, is 200 as, the ratio of the measured pulse width and the transform-limited pulse width is 0.97 in this case. The close match between the measured and transform-limited pulse widths indicates that all harmonics are locked in phase. It is also worth mentioning that the strongest and also the shortest attosecond pulse were obtained in the 40-torr case. Consequently, we were able to demonstrate the self-compression of the attosecond harmonic pulse without much loss in the intensity of the attosecond pulse.

The self-compression technique of the intrinsic attosecond pulses has some advantages over the other method involving the use of a metallic x-ray filter. Since the harmonic generation medium itself acts to compress the pulses, no other absorption loss was involved. This is a significant advantage since the metallic x-ray filter introduces large undesirable absorption due to oxide layers, which invariably form on the metal film. Further, continuous chirp control of the attosecond pulses can be easily achieved by adjusting the pressure of the harmonic generation medium. The self-compression technique discussed above can also be applied to the case of absorption-limited harmonic generation, provided the harmonic generation conditions are well optimized [15]. This will lead to the generation of strong, transform-limited attosecond pulses without sacrificing harmonic strength.

In summary, we have demonstrated the self-compression of attosecond pulses, whose intrinsic chirp was compensated by the harmonic generation medium itself, and have successfully generated nearly transform-limited attosecond pulses. Our results showed that, with argon as the generation medium, the chirp of the harmonics higher than the 25th order was well compensated. Other gases, such as Xe or Kr, can also be used for the chirp compensation since they have regions of negative GDD. In the case of Ne or He, the chirp compensation by the harmonic generation medium is not effective, and one needs to use two separate gas cells, one for generation and the other for the chirp compensation. When two gas cells are used, intrinsic harmonic chirp can be compensated over a wide spectral range due to the extra freedom of individually adjusting the gas pressure, and the duration of the compressed attosecond pulse may reach below 100 as.

This research was supported by the Korea Science and Engineering Foundation through the Creative Research Initiative Program.

Note added.—For the validity confirmation of the RABITT measurement, a comparison was made, under similar experimental conditions, with the frequency-resolved optical gating technique that was experimentally demonstrated recently by the authors for the full temporal characterization of an attosecond pulse train [16]. The agreement was better than 10%.

[1] M. Hentschel *et al.*, Nature (London) **414**, 509 (2001).
[2] Y. Mairesse *et al.*, Science **302**, 1540 (2003).
[3] K. T. Kim *et al.*, Phys. Rev. A **69**, 051805(R) (2004).
[4] S. Kazamias and Ph. Balcou, Phys. Rev. A **69**, 063416 (2004).
[5] A. Morlens *et al.*, Opt. Lett. **30**, 1554 (2005).
[6] R. López-Martens *et al.*, Phys. Rev. Lett. **94**, 033001 (2005).
[7] K. T. Kim *et al.*, Appl. Phys. B **79**, 563 (2004).
[8] P. B. Corkum, Phys. Rev. Lett. **71**, 1994 (1993).
[9] D. G. Lee *et al.*, Phys. Rev. A **63**, 021801(R) (2001).
[10] P. Salières *et al.*, Science **292**, 902 (2001).
[11] B. L. Henke *et al.*, At. Data Nucl. Data Tables **54**, 181 (1993).
[12] D. G. Lee *et al.*, Appl. Phys. Lett. **81**, 3726 (2002).
[13] P. M. Paul *et al.*, Science **292**, 1689 (2001).
[14] H. T. Kim *et al.*, Phys. Rev. A **69**, 031805(R) (2004); V. Tosa *et al.*, Phys. Rev. A **71**, 063807 (2005).
[15] E. Constant *et al.*, Phys. Rev. Lett. **82**, 1668 (1999).
[16] Chang Hee Nam *et al.*, in Conference on Lasers and Electro-Optics, Baltimore, Maryland, USA, 2007, Technical Digest, paper CTuW3.