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Attosecond chirp compensation over broadband high-order harmonics to generate near transform-limited 63 as pulses

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Abstract. By generating broadband high-harmonic pulses from neon and compensating for attosecond chirp by the material dispersion of argon, the generation of near transform-limited 63 as pulses was achieved. The spectral phase analysis showed that, without proper compensation, the attosecond chirp of the broadband harmonics caused splitting of attosecond high-harmonic pulses in addition to pulse broadening. Although it was attained only within a limited spectral range, the attosecond chirp compensation was successful in bringing out pulse compression over broad harmonics, which signifies the effectiveness of the attosecond chirp compensation by material dispersion.

High harmonics emitted from atoms driven by an intense femtosecond laser pulse is a new kind of light source with attosecond duration [1]. As the spectral bandwidth of high harmonics can be very broad with an equal spacing between adjacent harmonics, an attosecond pulse train can be generated. The high-harmonic pulses, however, contain inherent chirp originating from the harmonic generation processes [2], causing pulse broadening. In order to obtain transform-limited attosecond pulses, the attosecond chirp (called ‘atto-chirp’) has to be compensated. As the attosecond chirp of harmonics contributed from short trajectory components is positive, its compensation in a material with negative group-delay dispersion (GDD) was proposed [2] and realized using metallic [3, 4] and gaseous media [5]. As negative GDD can be found only in

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certain limited wavelength regions, the application of this concept to broad harmonic orders, needed for very short attosecond pulse generation, is yet to be proved.

In order to obtain transform-limited ultrashort pulses, two conditions of broad bandwidth and chirp-free spectral phase have to be satisfied. Since high-order harmonics can cover a broad spectrum, the first condition can be met by generating harmonic radiation over a wide spectral range [6]. High harmonics in the plateau region can form a frequency comb [7, 8], similar to a mode-locked femtosecond laser, that extends up to the harmonic cutoff frequency set by $3.17U_p + I_p$. Here, U_p is the ponderomotive potential, proportional to the driving laser intensity, and I_p is the ionization potential of the harmonic generation medium. A broad harmonic spectrum can be generated by applying intense laser pulses to a suitable gas medium. However, broader harmonic bandwidth does not automatically lead to shorter pulse duration due to the inherent atto-chirp contained in the harmonic pulses [2, 3]. As a consequence, the atto-chirp has to be removed in order to realize the shortest possible pulse duration. In this paper, we analyzed the spectral phase of broadband high harmonics generated from Ne and, after compensating for the chirp by the material dispersion of Ar, achieved the generation of 63 as pulse trains, close to the transform-limited value of 47 as.

The compensation of atto-chirp is crucial to produce transform-limited pulses. Several methods, such as the use of prism pairs, grating pairs or chirped mirrors, were applied for the chirp compensation of femtosecond laser pulses with the positive chirp gained while propagating through optical media [9, 10]. Although the high-harmonic pulses contributed from short trajectory components also have positive chirp structure [2, 11], the chirp compensation methods in the visible/infrared (IR) region cannot be applied easily to harmonic pulses due to strong absorption and poor reflectivity in the soft x-ray region. The use of chirped mirrors may still be attempted [12], but it is not a straightforward method because of the case-specific design and manufacturing difficulty. A different chirp compensation method is, thus, needed for atto-chirp compensation.

As a practical method to overcome this problem, atto-chirp compensation by material dispersion was proposed [2]. The positive atto-chirp of harmonic pulses can be compensated by utilizing a metallic foil with negative GDD, frequently used as an IR blocking x-ray filter, such as Al or Zr, because such a metallic foil has large negative GDD at the low-energy side of its x-ray transmission window. The atto-chirp compensation by adopting an Al x-ray filter was applied to the harmonics from Ar, where transform-limited 170 as pulses were obtained [4]. The same group also obtained 130 as pulses from Ne harmonics using Zr for chirp compensation [13]. The use of a gas medium for chirp compensation, instead of a metallic foil, was proposed because rare gases, generally used for harmonic generation medium such as Ar, Kr and Xe, have good transmittance and large negative GDD in certain wavelength regions. The chirp compensation with a gas medium is advantageous over that with a metallic filter because attosecond chirp can be finely controlled by adjusting the gas pressure, even during an experiment. Additionally, the large absorption loss due to the formation of oxide layers on a metallic filter can be avoided. By utilizing Ar as the chirp compensation medium, as well as the harmonic generation medium, transform-limited 206 as pulses were generated [5]. The atto-chirp compensation by material dispersion has, thus, been proven to be a practical method for the generation of transform-limited attosecond pulses.

For the generation of much shorter harmonic pulses, special consideration of broadband harmonic generation and proper atto-chirp compensation should be made. Broad-bandwidth high harmonics can be generated from Ne at strong laser intensity. At the applied laser intensity

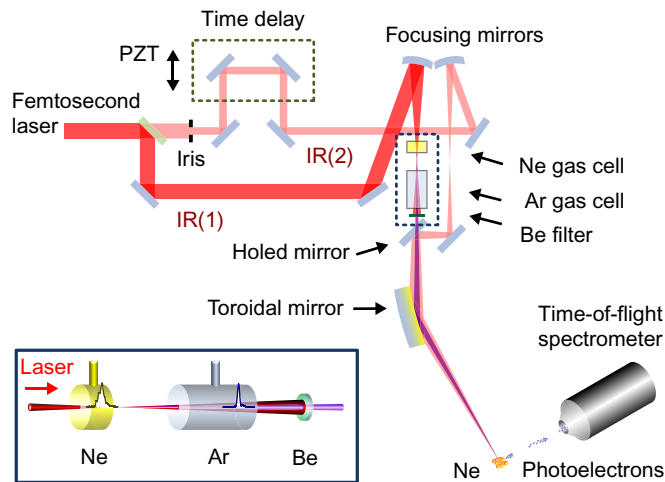


Figure 1. Schematic diagram of the experimental setup employing a two-gas-cell system for harmonic generation and atto-chirp compensation. The inset shows the geometry of the two-gas-cell system with a Be filter.

of $5 \times 10^{14} \text{ W cm}^{-2}$, the harmonic cutoff can be extended up to the 75th order. In order to deliver the broad harmonics to the interaction region, the x-ray filter, used to block IR laser pulses, should have sufficient transmission region, for which Be, instead of Al, was chosen [14]. Be has a reasonably good transmission from 40 to 112 eV, which is more than twice the bandwidth of Al. The broad harmonics, generated from Ne and filtered with Be, can cover over 25 harmonics and, consequently, can generate attosecond pulses shorter than 50 as. The broad harmonics, however, cannot form short attosecond pulses without proper atto-chirp compensation. Since it is not easy to find a material with negative GDD for the broad range, Ar with a large negative GDD from 38 to 60 eV was selected for atto-chirp compensation [5]. The efficient chirp compensation in the low-energy region may contribute to short pulse formation by flattening the variation of the spectral phase over a broad spectral range.

For the implementation of broad harmonic generation from Ne and atto-chirp compensation with Ar, a two-gas-cell system was adopted by separating the chirp compensation cell from the harmonic generation cell. A schematic diagram of the experimental setup is shown in figure 1. A 1-kHz Ti:sapphire femtosecond laser pulse was divided into two parts for harmonic generation [15] and for the temporal characterization of harmonic pulses using the reconstruction of attosecond beating by interference of two-photon transition (RABITT) method [16]. The first pulse, carrying 80% of the energy, was focused to the 3 mm Ne gas cell placed before the laser focus for strong harmonic generation through the guided propagation obtained by balancing the geometrical focusing and plasma defocusing effects of the laser beam [17, 18]. The Ne harmonics were carefully controlled by adjusting harmonic generation parameters, such as driving laser energy and chirp, gas pressure, position and medium length for strong and sharp peaks with even intensity and good phase relation between adjacent harmonics over a broad spectral region [19]. The harmonic pulse was then sent to the 9 mm Ar gas cell for atto-chirp compensation placed 5 cm after the Ne gas cell. The Ar gas cell was placed away from the laser focus so that no additional harmonics could be generated. The strong harmonics contributed from short trajectory components, existing in the central part of the harmonic beam [20, 21], were selected using an aperture (not shown) placed behind

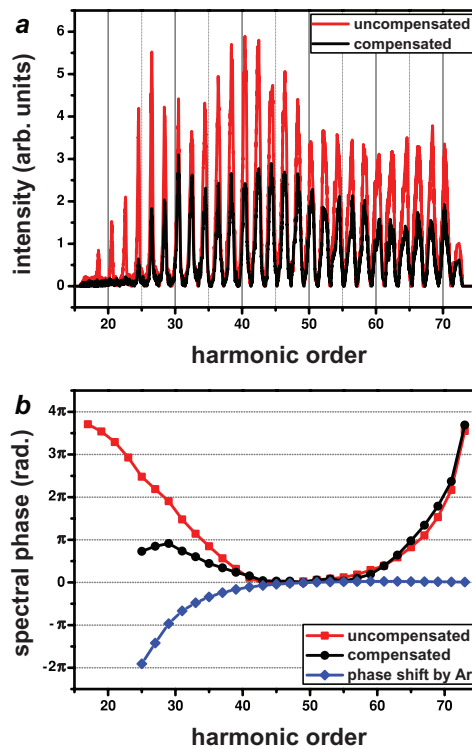


Figure 2. Harmonic spectra and spectral phases of uncompensated and compensated harmonic pulses. (a) Photoelectron spectra obtained without (black line) and with (red line) atto-chirp compensation are shown for the harmonics from the 17th to the 73rd order. The gas pressure in the Ar cell used for atto-chirp compensation was 25 Torr. The harmonics, lower than the 25th order, were strongly absorbed by Ar. (b) Spectral phases retrieved from the RABITT measurements for the uncompensated (red square) and compensated cases (black circle) are presented. The spectral phase shift gained while propagating through the 9 mm cell filled with 25 Torr Ar is also shown (blue diamond).

the Ar gas cell. In order to block the co-propagating IR laser pulse, a 300 nm Be filter was installed. For the RABITT measurement the second time-delayed laser pulse with an intensity of $2 \times 10^{11} \text{ W cm}^{-2}$ was combined with the harmonic pulse using a 1-mm-holed mirror. The high-harmonic pulse and the probe laser pulse with a time delay were focused together into an effusive Ne beam using a gold-coated toroidal mirror, thereby generating photoelectrons. The photoelectron spectra were measured using a magnetic-bottle-type time-of-flight (TOF) spectrometer that collected photoelectrons from Ne ionized by the harmonic and IR pulses. The measured energy resolution of TOF was 11 meV at 0.7 eV and, to reduce energy spreading at high photoelectron energy, photoelectron spectra for the harmonics with orders higher than the 47th order were taken with a bias voltage of 50 V. The photoelectron spectra consisted of harmonic and sideband signals modulated twice per optical cycle. The sideband modulation containing harmonic phase information originated from the interference of electrons ionized through different paths. The inset shows a magnified view of the two-gas-cell system used for harmonic generation and atto-chirp compensation.

High-harmonic pulses generated from Ne without chirp compensation were analyzed first. The harmonic spectrum obtained from the photoelectron energy spectrum is presented

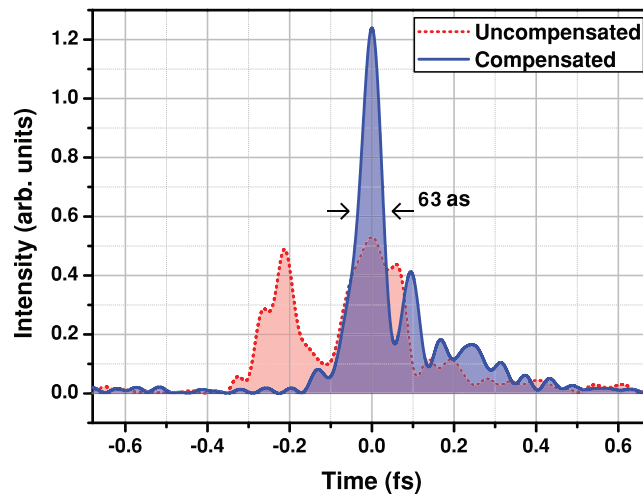


Figure 3. Reconstructed temporal profiles of harmonic pulses with and without atto-chirp compensation. The uncompensated harmonic pulse (red dotted line) shows a multi-peaked profile with broadened duration. The compensated harmonic pulse (blue solid line) shows a 63 as pulse, while the transform-limited value is 47 as.

in figure 2(a), showing the harmonics from the 17th to the 73rd order. These harmonics are broad enough to support a transform-limited duration of 45 as. From the RABITT measurement, the spectral phase of harmonic pulses was retrieved and, hence, the temporal reconstruction of attosecond harmonic pulses was achieved. As shown in figure 2(b), the spectral phase of Ne harmonics, presented in red squares, exhibits mainly a quadratic structure due to the inherent positive chirp of high-harmonic pulses. The reconstructed temporal profile in figure 3 shows a multi-peak profile with broadened duration. The duration of the central pulse is about 160 as, which is much longer than the transform-limited value. Even if the spectral bandwidth of the harmonic pulse increased significantly, the harmonic pulse duration was not shortened due to atto-chirp. The formation of the multi-peaked profile can be understood from the spectral phase analysis of the harmonic pulses. The first peak is formed by the harmonics from the 19th to the 45th order and the second peak comes from the harmonics from the 47th to the 61st order. It also shows that the difference in the group delay between the first and second peaks is 210 as, which matches well the temporal separation between the two peaks. In figure 3, the temporal profile of the uncompensated harmonic pulses clearly reveals these features. Consequently, it is clear that the broad harmonic generation did not shorten the harmonic pulse duration; rather it produced multi-peaked attosecond pulses with a broadened duration.

In order to optimize the harmonic pulse duration, atto-chirp compensation was performed by utilizing Ar. The temporal characterization of harmonic pulses was done while changing Ar pressure to find out the conditions for optimum chirp compensation. The shortest attosecond pulse was produced with an Ar pressure of 25 Torr. The harmonic spectrum and retrieved spectral phase of the compressed harmonic pulse are presented in figures 2(a) and (b), respectively. The transform-limited duration, corresponding to the harmonic spectrum, slightly increased to 47 as, since low-order harmonics were absorbed in Ar. Figure 2(b) shows that the spectral phase variation below the 41st order decreased dramatically because of the negative GDD of Ar in the region, indicating that the chirp compensation of harmonic pulses was quite

effective in the low-frequency region. The spectral phase shift gained while propagating through the 9 mm cell filled with 25 Torr Ar is also shown in figure 2(b), which matches well the spectral phase difference between the chirp-compensated attosecond pulse and the uncompensated attosecond pulse. The spectral phase in figure 2(b) shows that the chirp compensation is effective up to the 40th order; however, it shows that the spectral phase variation of the harmonic pulse is less than π up to around the 65th order. This means that a much larger number of harmonics can participate in the short pulse formation through coherent addition of harmonics than that expected from the range of chirp compensation by Ar. The RABITT analysis clearly showed that the broader participating harmonics resulted in the generation of 63 as pulses, as shown in figure 3, which corresponds to 1.2 optical cycles. The small satellite peaks in figure 3 appeared mainly from the harmonics of the higher orders that were strongly affected by the high-frequency edge of the Be transmission window with large positive GDD. It is noted that the peak intensity of the compensated case became stronger than that of the uncompensated case in figure 3, even though the harmonic intensity was reduced to about half of the uncompensated case due to better synchronization of harmonics. Consequently, near-transform-limited attosecond high-harmonic pulses, much shorter than the uncompensated case, were generated by obtaining effective atto-chirp compensation.

In conclusion, near-transform-limited 63 as high-harmonic pulses were produced by generating broad harmonics from Ne and compensating atto-chirp in Ar. This result showed that the effective number of harmonics participating in the short pulse formation is much larger than that expected from the range of chirp compensation and clearly demonstrated the effectiveness of atto-chirp compensation by material dispersion, especially in a gas medium. As other gases, such as Kr or Xe, have negative GDD in different wavelength regions, the atto-chirp compensation can be realized for harmonic pulses in a variety of wavelength regions. As it can be equally applied to isolated single attosecond pulses obtained using carrier-envelope-phase controlled few-cycle laser pulses [22], the atto-chirp compensation by material dispersion, especially of gaseous media, is a powerful and practical tool for both attosecond pulse trains and isolated attosecond pulses. The generation of near-transform-limited attosecond pulses will be a valuable tool to enhance temporal resolution in attosecond science for exploring ultrafast atomic and molecular dynamics.

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