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Single sub-50-attosecond pulse generation from chirp-compensated harmonic radiation using material dispersion

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A method for obtaining a single sub-50-attosecond pulse using harmonic radiation is proposed. For the generation of broad harmonic radiation during a single half-optical cycle, atoms are driven by a femtosecond laser pulse with intensity above the saturation intensity for optical field ionization and hence experience a large nonadiabatic increase of the laser electric field between optical cycles. Although the chirped structure of the harmonic radiation imposes a limit on the minimum achievable pulse duration, we demonstrate that its positive chirp can be compensated by the negative group delay dispersion of an appropriately selected x-ray filter material, used also for the spectral selection, resulting in a single attosecond pulse with a duration less than 50 as.

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High-order harmonics generated by atoms interacting with an intense femtosecond laser pulse can be a source of attosecond (1 as$=10^{-18}$ s) pulses. The fact that the regularly spaced harmonics resemble the spectrum of a mode-locked laser has prompted the suggestion that an attosecond pulse train can be obtained by selecting a range of harmonics. This was indeed confirmed in recent experiments [1–3]. The generation of a single attosecond pulse can be very important for applications in which the pulse duration determines the temporal resolution. By selecting the harmonic radiation (implying all the radiation including a broad continuum) emitted only at the peak of a femtosecond laser pulse, Hentschel *et al.* [4] demonstrated the generation of a single attosecond pulse of 650 as. For a significant reduction in the duration of the single attosecond pulse, two limitations have to be overcome: generation of a broad frequency bandwidth and ensuring a linear phase relation over the selected frequency range. In the case of generating an attosecond pulse train, harmonic radiation can support such a broad bandwidth [5,6], but the chirped structure of harmonics seriously restricts the lowest achievable pulse duration [3].

In this Rapid Communication, we propose a method for obtaining a single sub-50-as pulse using chirp-compensated harmonic radiation. The continuum radiation emitted at the peak of a femtosecond pulse is not broad enough for supporting a very short single attosecond pulse ($<$100 as). Hence as the first step we propose to obtain a broad continuum radiation from atoms by subjecting them to a large nonadiabatically increasing electric field over successive half optical cycles. Though the resulting spectral width of the continuum is broad enough for the generation of an attosecond pulse much shorter than 100 as, the frequency chirp contained in the continuum radiation severely restricts the attainable attosecond pulse duration. As the second major step we demonstrate that the positive chirp, inherently contained in the continuum radiation, can be compensated by the negative group delay dispersion (GDD) of an appropriately selected x-ray filter material.

Harmonic radiation can be used for the generation of either an attosecond pulse train or a single attosecond pulse. Harmonic radiation is, in general, emitted during every half optical cycle and forms a train of attosecond pulses. The generation of a single attosecond pulse, on the other hand, requires the use of a part of the harmonic radiation emitted during a particular half cycle, which is naturally a continuum radiation. Since the highest frequency (or the cutoff order) of harmonics emitted during each half cycle increases until the pulse peak is reached, when the laser intensity is lower than the saturation intensity, a single attosecond pulse can be produced by selecting the continuum radiation emitted only during the half cycle around the pulse peak (here we assume a cosine pulse shape) [7]. However, the spectral range of the continuum radiation in the cutoff region is still not broad enough to generate very short pulse (say, below 100 as), since the electric field variation between adjacent optical cycles near the pulse peak is not large enough.

In our approach we propose the application of an intense carrier-envelope-phase- (CEP-) stabilized femtosecond laser pulse having an intensity above the saturation intensity. In this case harmonic generation occurs only at the leading edge of the laser pulse since most neutral atoms are ionized before the pulse peak is reached. The electric field variation in the leading edge of the laser pulse is larger than that around the pulse peak [8]. In the case of a Gaussian laser pulse, the relative electric field variation from one optical cycle to the next increases linearly as time goes negative; time is defined to be zero at the pulse peak. By applying a few-cycle laser pulse, with an intensity above the saturation intensity, we can obtain harmonic radiation containing a broad continuum, since harmonic radiation originates from atoms experiencing large nonadiabatic increase of the electric field at the leading edge of the laser pulse. We then have to examine the phase variations of the selected continuum radiation. As the spectral structure of the harmonic radiation reflects the rapid increase of the instantaneous electric field during the half

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cycle, its spectral phase inherently contains a chirped structure [9] and, hence, only a limited range of the continuum radiation can be coherently added. We then demonstrate that the chirp of the continuum radiation can be removed by using an appropriately chosen x-ray filter, thereby producing a single attosecond pulse of duration less than 50 as.

Harmonic radiation from neon atoms, exposed to a 5-fs, 800-nm laser pulse with peak intensity of 4.7×10^{15} W/cm^2, which is greater than the saturation intensity, is obtained by solving the time-dependent Schrödinger equation (TDSE) coupled with Maxwell equations. Here the one-dimensional propagation calculation is performed taking into account the focusing geometry and the self-phase modulation of the propagating laser pulse in an ionizing medium [10]. The neon target medium, of 0.5 mm length and 5 Torr pressure, is placed 10 mm beyond the laser focus so that only harmonics of short trajectories survive after propagation [11,12]. The time-frequency structure of the harmonic radiation, calculated using the spectrogram method [13], is shown in Fig. 1(a). The semiclassical calculations containing only the short trajectory components (obtained without considering ionization), indicated as dotted lines in Fig. 1(a), match well with the TDSE results. At this laser intensity, almost all neutral neon atoms are ionized before the pulse peak ($t=0$) is reached and the dominant harmonic radiation is generated near $t=-1.0$ (cycle). The selection of continuum radiation, with frequencies larger than $100\omega_0$ (ω_0=laser frequency), leads to the generation of a single attosecond pulse since it constitutes the radiation generated only within the half cycle at $t=-1.0$ (cycle).

On the other hand, when a laser intensity lower than the saturation intensity is applied, the spectral range of the continuum radiation is much narrower. In this case the highest harmonic frequency is obtained at the peak of the laser pulse. Figure 1(b) shows the harmonic radiation calculated for the same conditions as in Fig. 1(a), except that the peak laser intensity is 7.0×10^{14} W/cm^2 and the medium length is 1 mm. In this case the selectable spectral width of the continuum radiation for a single attosecond pulse generation is reduced to about $10\omega_0$, which would allow a pulse duration of only 120 as even if the entire selected radiation were coherently added. The relative electric field variation between successive half optical cycles at the time of strong harmonic radiation is 16% in the case of Fig. 1(b), while it is 70% in the case of Fig. 1(a). Thus, the advantage of applying a laser intensity higher than the saturation intensity for generating a broad continuum spectrum is clearly evident. It may also be mentioned that the choice of a higher laser intensity enables us to select the continuum bandwidth in the region of maximum slope of the curve in Fig. 1(a); in this region the spectral phase variation is small as discussed below.

For the generation of a single attosecond pulse with duration as short as possible, the phase relation of the broad continuum radiation, obtained by applying the intense 5-fs laser pulse [Fig. 1(a)], has to be estimated. During a half optical cycle, the frequency of the harmonic radiation increases rapidly in time, being positively chirped. This is inevitable in all harmonic radiation sources pumped by a laser pulse. Here we select the harmonic radiation in the midfrequency region of Fig. 1(a), characterized by linear frequency

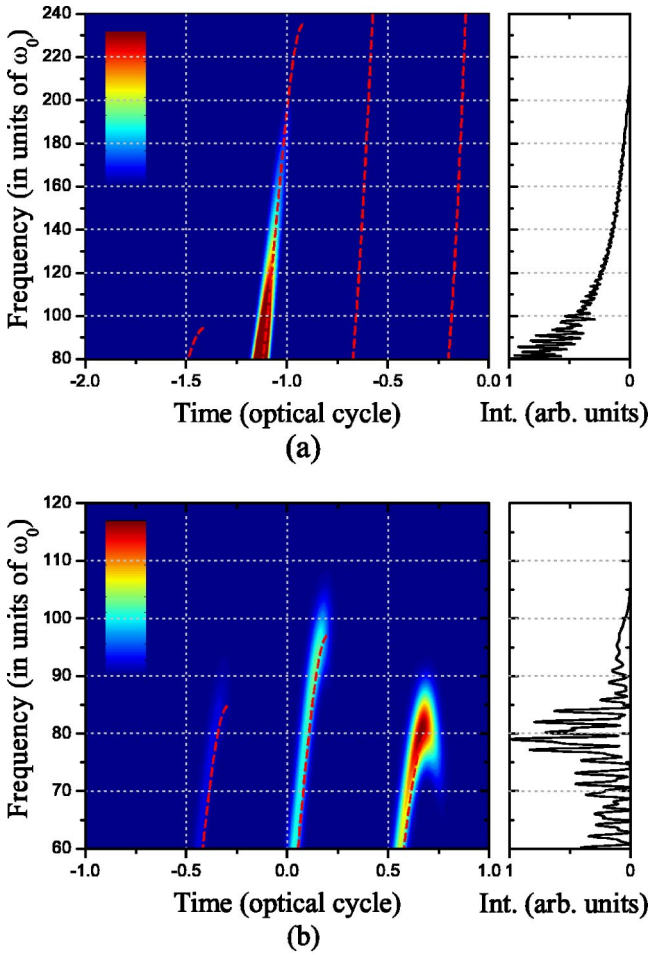

FIG. 1. (Color online) Spectrogram of the harmonic radiation from neon atoms exposed to 5-fs, 800-nm pulse with peak intensity (at the medium) of (a) 4.7×10^{15} W/cm^2 and (b) 7.0×10^{14} W/cm^2. The dotted lines represent the short-trajectory component of the harmonic radiation obtained from the semiclassical calculation. The figures on the right side of the spectrogram show the harmonic spectra.

chirp. In this case the spectral phase varies quadratically with frequency and we may express the phase of an attosecond pulse in the frequency domain as

$$\tilde{\phi}_h(\omega) \simeq \frac{1}{2\alpha}(\omega-\omega_c)^2, \tag{1}$$

where α is the chirp coefficient at the center (ω_c) of the selected frequency region and is simply the slope of the curve shown in Fig. 1(a). Even though α is large enough to make the phase variation with frequency small, the quadratic phase variation can be large enough to destroy a constructive addition of different frequency components. Assuming that we use a Gaussian transmission filter for the frequency selection, the maximum bandwidth required for minimum pulse width is given by $\Delta\omega_{\max}=\sqrt{4(\ln 2)\alpha}$. If the bandwidth of the transmission filter is $\Delta\omega$ in full width at half maximum (FWHM), the pulse width of the resulting short pulse passing through the filter is given by

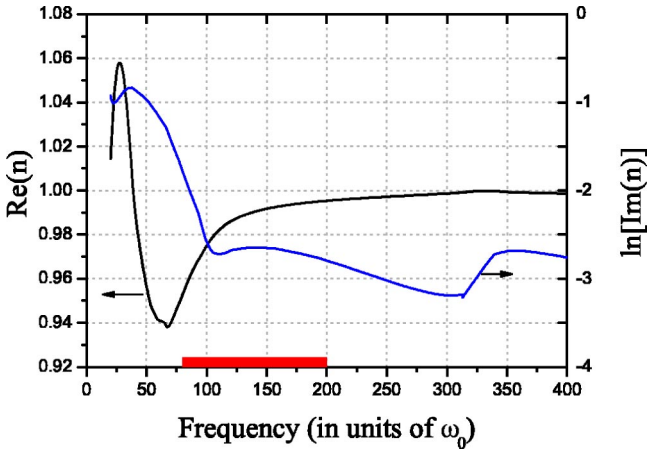

FIG. 2. (Color online) Real and imaginary parts of the refractive index of a Sn filter. The spectral range of negative GDD is marked as a thick red line.

$$\tau_{FWHM} \simeq \frac{4\ln 2}{\Delta\omega}\sqrt{1+\left(\frac{\Delta\omega^2}{4(\ln 2)\alpha}\right)^2}. \qquad (2)$$

For the case of Fig. 1(a), α is 860 fs^{-2} and $\Delta\omega$ is $21\omega_0$, which results in a pulse width of 80 as. Although the accommodation of broad harmonic radiation is essential for the generation of a short attosecond pulse, the bandwidth over which the radiation gets constructively added is limited as long as the chirp exists.

For the compression of the chirped harmonic pulse, the chirp contained in the pulse should be minimized. The positive chirp in the harmonic radiation can be removed using a material with a negative GDD. We propose the use of a judiciously selected x-ray filter which serves to compensate the chirp of the harmonic pulse in addition to the selection of the useful spectral range. The material dispersion of an x-ray filter modifies the phase relation of an attosecond pulse propagating through it. An x-ray filter with a "flat-top" transmission window that has a negative GDD near the low-frequency side of the transmission window could be best suited for the chirp compensation and spectral selection at the same time. In general, materials belonging to the fifth row of the periodic table have such a flat-top transmission window over the energy range between the *M* and *N* absorption edges [14].

We now show how the x-ray filter compensates the positive chirp of the harmonic radiation by considering the dispersion properties of an x-ray filter material. In order to understand the effect of the filter, we need to look into the frequency-dependent complex refractive index given as

$$n(\omega) = 1 - \delta(\omega) + i\beta(\omega), \qquad (3)$$

where δ accounts for the phase shift in the material and β is chosen positive for describing the absorption in the material. They are related by the Kramers-Kronig relation so that the real part of the refractive index can be calculated from the photoabsorption data [14].

Figure 2 shows the real and imaginary parts of the refractive index of an Sn(Tin) filter plotted against the frequency.

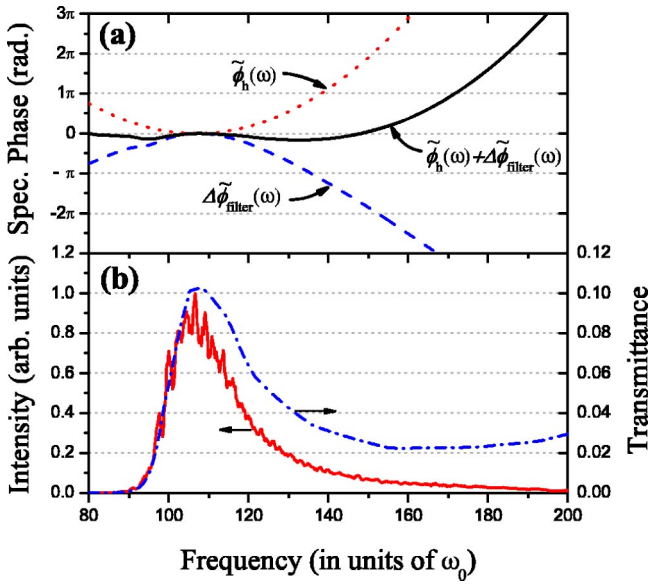

FIG. 3. (Color online) (a) Chirp compensation of positively chirped harmonic radiation by a Sn x-ray filter. The spectral phase of the harmonic radiation with a positive linear chirp centered at $107\omega_0$, $\tilde{\phi}_h(\omega)$, and the spectral phase shift induced by a 700-nm-thick Sn filter, $\Delta\tilde{\phi}_{\text{filter}}(\omega)$, are shown, respectively, by dotted and dashed lines. The spectral phase of the harmonic radiation after the Sn filter is shown as the solid line. (b) Spectral intensity of the harmonic radiation corresponding to Fig. 1(a), transmitted through the Sn filter (solid line) and the transmittance of the Sn filter (dash-dotted line).

The imaginary part of the refractive index (and also transmission) drops sharply starting from $40\omega_0$ up to $100\omega_0$ and remains fairly constant up to $310\omega_0$. We note that the Sn filter has a negative GDD in the region between $80\omega_0$ and $200\omega_0$ (marked by the thick red line in Fig. 2). Since this kind of transmission window is not unique to Sn, we may choose other materials for x-ray filters having a similar transmission window, such as Zr, Ag, or In, depending on the frequency range of interest. Consequently, we can compress any positively chirped harmonic pulse by using a carefully selected x-ray filter originally used for spectral selection.

For the case corresponding to Fig. 1(a), we choose an Sn filter for the spectral selection and chirp compensation. After the selection of an x-ray filter material with a suitable transmission window and dispersion property, we can compute the thickness of the filter to achieve optimum chirp compensation, retaining, at the same time, sufficient transmission. The spectral phase shift induced by the x-ray filter of thickness z is then given by $\Delta\tilde{\phi}_{\text{filter}}(\omega) = -\omega\delta(\omega)z/c$. The quadratic spectral phase of the linearly chirped harmonic pulse can be compensated reasonably well using a 700-nm-thick Sn filter. The spectral phase of the attosecond pulse obtained from Eq. (1) and the spectral phase shift $\Delta\tilde{\phi}_{\text{filter}}$ induced by the 700-nm-thick Sn filter are shown in Fig. 3(a). The total spectral phase variation, also shown in Fig. 3(a), with the Sn filter included, is much less than π over the FWHM of the transmission window of the Sn filter. The 700-nm-thick Sn filter, shown in Fig. 3(b), has 10% transmittance at the center frequency of transmittance and higher than 5% throughout

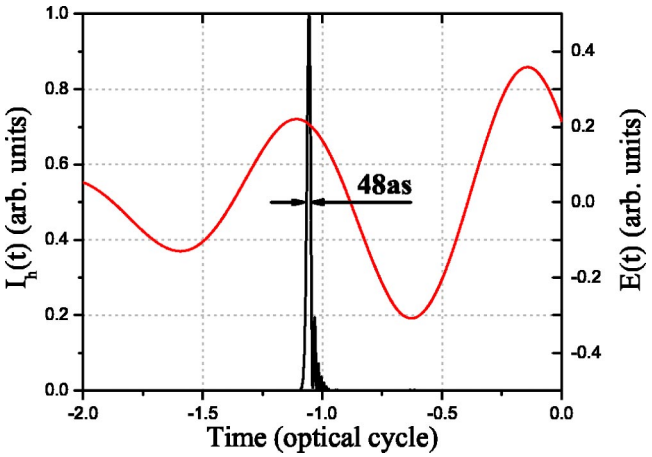

FIG. 4. (Color online) Temporal profile of the chirp-compensated attosecond pulse in the 700-nm Sn x-ray filter is shown along with the electric field of the 5-fs driving laser pulse.

its transmission window from $100\omega_0$ to $126\omega_0$. The spectral intensity of harmonics passing through the Sn filter is also shown in Fig. 3(b). As a result, the chirp-compensated harmonic radiation can be constructively added to generate a short single attosecond pulse.

Figure 4 shows the final pulse shape of the chirp-compensated harmonic pulse transmitted by the Sn filter. The pulse duration of the single harmonic pulse is 48 as, considerably shorter than the 89-as pulse obtained without considering the material dispersion. The transmitted harmonic spectrum shows a long high-frequency tail with poor chirp compensation. This causes a ripple in the tail part of the single attosecond pulse, which may be removed by using an additional x-ray filter. The contrast ratio of the 48-as pulse to the one generated during the previous half optical cycle is larger than 1000, mainly due to the application of a strong nonadiabatically increasing laser electric field on the atoms.

Figure 4 also shows the temporal profile of the laser electric field. In our analysis, we have assumed the laser field at the focus as a cosine function with a CEP value of 0; the CEP shift seen in Fig. 4 is due to the Gouy phase shift at the target position. The CEP jitter leads to a temporal variation of the laser pulse shape and, hence, varies the generation time of the attosecond pulse with respect to the peak of the laser pulse envelope ($t=0$). However, the CEP jitter of 0.1 rad, obtainable with a CEP locking method [15], does not cause a significant change in the results shown in Fig. 4. In addition, the frequency bandwidth of the continuum radiation depends on CEP, which can be neglected in the conditions of Fig. 1(a) because it is only about $\pm 2\omega_0$ for the above CEP jitter. Further, the spatial distribution of the laser intensity should be considered for more rigorous analysis, since it causes position-dependent attosecond pulse generation. However, this effect may be minimized by utilizing the profile-flattening technique, which can provide a nearly uniform laser intensity distribution [16].

In summary, we have demonstrated a method to generate a single sub-50-as pulse by employing a 5-fs laser pulse with intensity above the saturation intensity for the generation of broad continuum radiation and by compensating the harmonic chirp by using a properly selected x-ray filter material. We have shown that the positive chirp contained in the broad continuum radiation can be compensated by an x-ray filter with a negative GDD in the spectral region of interest. The present calculations indicate that a single 48-as pulse can be generated under experimentally achievable conditions. We expect that this technique would become a valuable tool for generating a single attosecond pulse with a duration less than 50 as.

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