Generation of sub-10 fs ultraviolet Gaussian pulses

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A technique is proposed for generating sub-10 fs deep ultraviolet (DUV) pulses with Gaussian temporal and spectral profiles. In this approach, a broadband DUV pulse with a negative frequency chirp is generated and compressed by normal group-velocity dispersion in a transparent medium. The pulse shape is not significantly distorted by high-order spectral phase distortion. In principle, nearly transform-limited sub-6 fs UV pulses with pulse energies of several tens of microjoules are expected to be generated. Numerical simulations indicate that a Ti:sapphire chirped-pulse amplifier with a pulse energy of several millijoules and a pulse duration of 35 fs can be used to generate high-energy sub-6 fs UV pulses. The technique is partially demonstrated by an experiment using a low-power Ti:sapphire chirped-pulse amplifier to generate low-energy sub-10 fs single DUV pulses with waveforms that quantitatively agree with those predicted by the numerical simulations. This technique can be used to generate high-energy sub-10 fs DUV pulses with Gaussian temporal and spectral profiles for ultrafast spectroscopy in the gas phase as well as the liquid and solid phases. © 2010 Optical Society of America 320.7110, 320.5520. OCIS codes:

1. INTRODUCTION

Intense ultrashort laser pulses with pulse lengths shorter than 10 fs in the near-infrared (NIR) and visible regions are currently generated by self-phase modulation (SPM) in a hollow fiber filled with a rare gas [1-3]. The use of chirped mirrors in the NIR and visible regions has enabled intense few-cycle pulses to be generated with pulse energies of several hundred microjoules or even higher [1,3]. However, chirped mirrors for near-ultraviolet (NUV) pulses have oscillations in their negative group-delay dispersion (GDD) curves. This makes it difficult to compress NUV pulses without generating subpulses in the compressed temporal intensity profile due to high-order spectral phase dispersion, especially when the NUV pulses have a broad spectral width and support sub-10 fs transform-limited pulse durations [4–6]. No chirped mirrors are available for deep UV (DUV) pulses. Instead, grating compressors, prism compressors, and deformable mirrors have been utilized to compress DUV pulses [7–11]. The experimental setup for compressing DUV pulses is considerably more complex than that for compressing visible pulses. Furthermore, using a grating or a prism compressor induces high third- and higherorder dispersions, resulting in subpulses being generated in the compressed temporal profile. Third-order dispersion and GDD can be simultaneously compensated for by employing a prism and grating compressors [12], but this technique requires a more complex setup and substantially reduces the output pulse energy. High-order dispersion can be compensated for by using a deformable mirror, but doing so may introduce spatial chirp [8]. Moreover, satellite pulses have been observed in compressed temporal profiles [8], and this technique has not been used to generate pulses shorter than 7 fs.

Sub-10 fs [13] and sub-4 fs DUV pulses [14] have been generated in vacuum chambers. These pulses are suitable for ultrafast spectroscopy in the gas phase of a sample placed in the vacuum chamber. When using sub-10 fs pulses for ultrafast spectroscopy of solid and liquid samples, which involves using short pulses in an atmospheric environment, it is necessary to compensate for pulse broadening due to group-velocity dispersion (GVD) in air. This requires the use of a pulse compressor, so the aforementioned problems must be considered.

Chirped-pulse four-wave mixing (CP-FWM) can generate high-energy ultrashort DUV pulses. It does not require a pulse compressor and performs precise compensation without inducing appreciable high-order spectral phase distortion [15,16]. CP-FWM generates a negatively chirped DUV pulse; this pulse is compressed by material dispersion in a transparent medium. This method is similar to the technique that generates negatively chirped DUV pulses by frequency doubling a negatively chirped visible pulse [17]. DUV pulses with pulse durations of about 30 and 18.7 fs can be generated by CP-FWM and frequency doubling, respectively [17]. CP-FWM can be used to generate sub-10 fs UV pulses if a sub-10 fs idler pulse is first prepared [18]. Since the CP-FWM utilizes a broadband idler pulse, we refer to it as broadband CP-FWM (BCP-FWM). In BCP-FWM, a transform-limited sub-10 fs idler pulse with a Gaussian or sech² (i.e., smooth) spectral shape with no satellite pulses is prepared, and it is then chirped using a glass block with a normal GVD. It is essential to prepare an idler pulse with the aforementioned characteristics to generate ultrashort UV pulses with spectral and temporal profiles that are described by a Gaussian or sech² function. A UV pulse with a Gaussian spectral profile is suitable for ultrafast spectroscopy in the spectral range since its smooth spectral shape with no missing spectral components in the spectral bandwidth of interest should enable the wavelength and temporal dependences of the absorbance in ultrafast spectroscopy to be measured with high accuracy [19,20]. The temporal profile of UV pulses should also have a single-pulse shape for ultrafast spectroscopy, otherwise the pre- and postpulses (subpulses or satellites) will excite the sample, which can drastically alter ultrafast phenomena observed by spectroscopy. However, it is not easy to generate sub-10 fs transform-limited Gaussian or sech² NIR pulses using a Ti:sapphire chirped-pulse amplifier. Moreover, theoretical calculations predict that ultrashort UV pulses generated by BCP-FWM will have strong tails and will not be single pulses [18]. Thus, BCP-FWM requires further development to generate single sub-10 fs pulses suitable for use in ultrafast spectroscopy.

In this article, a method for generating sub-10 fs DUV pulses with Gaussian spectral and temporal profiles is proposed and theoretically investigated. This technique is a development of BCP-FWM, but it does not require sub-10 fs idler pulses; instead, it employs a high-energy 35 fs Ti:sapphire chirped-pulse amplifier. A 35 fs NIR pulse is spectrally broadened by SPM, and then used as the idler for BCP-FWM. Although the self-phase-modulated idler has a complicated spectral shape, the signal pulse generated by this BCP-FWM technique has a smooth spectral shape. Furthermore, the signal pulse has a single-pulse temporal profile. The smooth spectral structure and the single-pulse shape of the compressed signal pulse is obtained by temporal gating of the positively chirped component in the self-phase-modulated idler by using a pump pulse that is shorter than the idler. Temporal gating is realized by controlling the ratio of the pump-pulse duration to the broadband-idler pulse duration and by prechirping the idler pulse before it is spectrally broadened by SPM, which is necessary for generating DUV pulses with high-quality spectral and temporal profiles. Numerical simulations are utilized to investigate how the developed BCP-FWM technique generates sub-10 fs DUV pulses with Gaussian spectral and temporal shapes and pulse energies of several tens of microjoules. Quantitative agreement was found between the pulse shapes of DUV pulses obtained in the numerical simulation and those in a recent experiment that generated sub-10 fs pulses [21]. This result partially demonstrates the principle of the technique proposed in this paper.

The paper is organized as follows. The concept behind the proposed BCP-FWM technique is first discussed. A numerical simulation is performed, which quantitatively confirms the theory as a result of good agreement with recent experimental observations [21]. Numerical calculations predict that, without substantial alteration, the technique can be extended to generate 5 fs pulses with Gaussian spectral and temporal profiles. The advantages and the potential of this technique as well as its limitations are then discussed and the conclusions of the study are presented.

2. THEORY

In a hollow fiber filled with rare gas, perfect phase matching for FWM is satisfied at low gas pressures [7]. A low gas pressure may result in the pump, idler, and generated signal pulses having negligibly small GVDs. For negligibly small depletion of the pump-pulse energy, the amplitude of the signal pulse e_s generated by FWM is expressed as [22]

$$\varepsilon_s(t,z) = (zn_2\omega_s/cA_{\text{eff}})A(t)_p^2 A(t)_i \exp[i(2\phi_p(t) - \phi_i(t) + \pi/2)],$$
(1)

where n_2 is the nonlinear refractive index, ω_s is the signal carrier frequency, c is the speed of light in a vacuum, and $A_{\rm eff}$ is the effective core area of the hollow fiber [22]. A_p and A_i are the real amplitudes of the pump and the idler, respectively, and φ_p and φ_i are the phases of the pump and the idler, respectively. In the proposed BCP-FWM technique, a broadband NIR signal pulse is prepared by SPM in a gas-filled hollow fiber. The temporal profile of this self-phase-modulated laser pulse contains down-chirps as well as an up-chirp [22]. Only the up-chirp component is used to generate a linearly (negatively) chirped signal pulse [15,16]. This produces a signal pulse with a smooth spectral shape and with no high-order phase distortion. This is realized by making the pump pulse shorter than the self-phase-modulated idler for temporal gating the up-chirp component of the idler during BCP-FWM (Fig. 1). The temporal width of the up-chirp component is determined by the pulse duration of the idler before SPM. For this reason, the idler is prechirped to increase its pulse duration before the SPM process. The pulse duration of the idler pulse could also be broadened by propagating the idler pulse through a transparent medium after the SPM process; however, this does not increase the temporal width of the up-chirp component, which is critically important in the proposed technique. In this technique, the temporal width of the up-chirp component in the idler must be long since it determines the maximum pulse width of the pump pulse, which must be sufficiently long to induce a large negative chirp in the signal pulse to enable the signal pulse duration to be easily compressed by normal dispersion in a transparent medium.

3. NUMERICAL MODEL

To discuss BCP-FWM quantitatively, it is necessary to introduce coupled equations for nonlinear propagation in a hollow fiber. In the FWM process, the amplitudes of the pump (ϵ_p), idler (ϵ_i), and signal (ϵ_s) vary along the propagation axis (the *z* axis) in accordance with [22,23]

$$\partial \varepsilon_p / \partial z = i D_p \varepsilon_p + i (\omega_p / c) n_2 T_p \{ [|\varepsilon_p|^2 + 2|\varepsilon_i|^2 + 2|\varepsilon_s|^2] \varepsilon_p + 2\varepsilon_p^* \varepsilon_i \varepsilon_s \exp(i\Delta\beta z) \},$$
(2)



Fig. 1. (Color online) Scheme for temporal gating of the up-chipp component of the self-phase-modulated idler for generating a linearly chipped signal with a Gaussian spectrum. UC and DC represent upchipp and down-chip, respectively. A down-chipp signal is generated by the interaction between a down-chipp pump (NUV) and the upchipp component of a self-phase-modulated idler (NIR).

$$\begin{aligned} \partial \varepsilon_{i,s} / \partial z &= i D_{i,s} \varepsilon_{i,s} + i (\omega_{i,s}/c) n_2 T_{i,s} \{ [|\varepsilon_{i,s}|^2 + 2|\varepsilon_p|^2 \\ &+ 2|\varepsilon_{s,i}|^2] \varepsilon_{i,s} + \varepsilon_p^2 \varepsilon_{s,i}^* \exp(-i\Delta\beta z) \}, \end{aligned}$$
(3)

$$D_{k} = -\alpha_{k}/2 - (1/v_{k} - 1/v_{\text{signal}})(\partial/\partial t) - i(\beta_{k}^{(2)}/2)(\partial^{2}/\partial t^{2}) + i(\beta_{k}^{(3)}/6)(\partial^{3}/\partial t^{3}) + \cdots,$$
(4)

where β_k is the propagation constant inside the hollow fiber, asterisks indicate complex conjugates, ω_k is the angular frequency of the electric fields, and n_2 is the nonlinear refractive index of the core medium; the phase mismatch is given by $\Delta\beta = \beta_i + \beta_s - 2\beta_p$, and the higher-order dispersion is $\beta_k^{(n)} = \partial^n \beta / \partial \omega^n |_{\omega = \omega_k}$. The subscript k in these equations indicates the pump, signal, and idler. $T_k = \{1 + (i/\omega_k)(\partial/\partial t)\}$ contains the effect of self-steeping [22,23], and v_k is the group velocity of each wave. The coupled equations are expressed in the frame of reference propagating with the signal group velocity v_{signal} . The field amplitude ε_k is normalized such that $|\varepsilon_k|^2/A_{\rm eff}$ is equal to the field intensity in units of watts per square centimeter. Photoionization of the medium is not included in the wave equations since it does not appreciably affect the FWM process; this is because the interacting pulses are chirped and have low intensities and the low gas pressure inside the hollow fiber ensures that the phase-matching condition ($\Delta \beta = 0$) is satisfied.

In order to model the spectral broadening of the idler pulse before the FWM process, the photoionization also has to be taken into account due to the much higher gas pressure [24,25]:

$$\begin{aligned} \partial \varepsilon / \partial z &= iD\varepsilon + i(\omega/c)n_2 T |\varepsilon|^2 \varepsilon \\ &- \{i(\beta/2n^2\rho_c)T^{-1}\rho + \sigma\rho/2 \\ &+ UW(|\varepsilon|^2)(\rho_{nt} - \rho)/2|\varepsilon|^2\}\varepsilon, \end{aligned}$$
(5)

where ρ is the electron plasma density [25], ρ_c is the critical plasma density, ρ_{nt} is the density of neutral atoms, $W(|\varepsilon|^2)$ is the ionization rate of the gas medium, n is the refractive index, and σ is the inverse bremsstrahlung cross section. The plasma density ρ varies with time according to [25]

$$\partial \rho / \partial t = W(|\varepsilon|^2)(\rho_{nt} - \rho) + (\sigma/U)\rho|\varepsilon|^2.$$
(6)

The following values are used in the simulation: $\rho_c =$ $1.73 \times 10^{21} \ p \ {\rm cm}^3$ [25], $\rho_{nt} = 2.7 \times 10^{19} \ p \ {\rm cm}^{-3}$, and $\sigma =$ 1×10^{-19} cm², where p is the ratio of the gas pressure to atmospheric pressure [24,25]. The ionization potential U is 16 eV for argon and 14 eV for krypton. The ionization rate $W(|\varepsilon|^2)$ is $\sigma^K |\varepsilon|^{2K}$, where the multiphoton ionization cross section is $\sigma_K = 5.3 \times 10^{-125} \text{ s}^{-1} \text{ cm}^{2K} \text{ W}^{-K}$ for krypton and $K = U/\hbar\omega$ [26]. The nonlinear refractive index for argon in the NIR region is widely accepted to be $1 \times 10^{-19} p \text{ cm}^2/\text{W}$ [25,27,28], whereas it has been reported to be $2.9 \times 10^{-19} \ p \ \text{cm}^2/\text{W}$ at 248 nm [29]. This value for 248 nm is not consistent with the experimentally observed spectral broadening by SPM [30], and thus is viewed with uncertainty, as discussed in [31]. The value of n_2 for argon is expected to be of the order of $10^{-19} p \text{ cm}^2/\text{W}$ throughout the wavelength range from NIR to DUV. In the following discussion, n_2 of argon is taken to be $1 \times 10^{-19} p \text{ cm}^2/\text{W}$ [25,27,28] to simulate nonlinear pulse propagation in the presence of SPM and FWM at all wavelengths of the pump, signal, and idler. Numerical simulations

using this value of n_2 quantitatively reproduce our experimental data very well (see Section 4). This agreement is considered to be due to the small SPM effect induced by a small Kerr effect during the nonlinear propagation of BCP-FWM because of the low gas pressure and low peak intensities of the input pulses. In the NIR wavelength region, n_2 of krypton is taken to be $2.78 \times 10^{-19} p \text{ cm}^2/\text{W}$ [28]. The GVD in the rare gas-filled hollow fiber is calculated using the refractive index dispersions of argon and krypton reported in [32] and the waveguide dispersion. The coupled equations are solved using the split-step Fourier method [22].

4. RESULTS AND DISCUSSION

The experimental parameters given in [21] are used in the BCP-FWM simulation, and the schematic of the experimental scheme is shown in Fig. 2. A slightly negatively chirped 35 fs Gaussian pulse (transform-limited pulse width, 35 fs; center wavelength, 800 nm) is assumed to transmit through fused silica plates before being coupled into a Kr-filled hollow fiber (core diameter, 250 μ m; length, 600 mm), resulting in an input pulse with a duration of 52 fs and a pulse energy of 230 μ J. The pulse shape of the pump pulse generated by passing another 35 fs transform-limited pulse through a 200 μ m thick beta barium borate (BBO) crystal is calculated using coupled equations for second-harmonic generation [33] and a secondorder susceptibility of 2.74×10^{-12} m/V together with the GVD parameters of BBO. The temporal shape of the pump pulse, which is negatively chirped using a double-pass prism compressor, is then calculated using the model reported by Fork et al. [34,35] (although more rigorous equations are employed in this study than in [34,35]). The pump pulse coupled into the hollow fiber for BCP-FWM has a pulse duration of 116 fs.



Fig. 2. (Color online) Schematic of the experimental scheme.

Coupled pulse energies of 44 and 66 μ J are used for the idler and pump pulses, respectively; these values were estimated based on experimental pulse energies of the pump and idler pulses measured before and after the hollow-fiber chamber [21] and the calculated loss constant of the hollow fiber (core diameter, 140 μ m; length, 570 mm) [36]. A time delay between the two pulses that produces temporal overlap between the center regions of the idler and the pump pulse was used in the BCP-FWM simulation.

Figure 3 shows the results of the simulation and those of the corresponding experiment. For the simulation, the chirp rate (GDD) of the idler pulse focused into the first hollow fiber (to spectrally broaden the idler) was estimated in the following manner. The pulse duration and spectral width of the output pulse (with a transform-limited pulse duration of 35 fs) emerging from the chirped-pulse amplifier were used; that is, a transform-limited Gaussian pulse with the same pulse duration (35 fs) was assumed for the simulation. The material dispersion due to the transparent optical components between the chirped-pulse amplifier and the hollow fiber was calculated to estimate the chirp rate of the Gaussian pulse at the entrance of the hollow fiber. However, using the estimated chirp rate in the simulation resulted in a self-phase-modulated idler spectrum [broken curve in Fig. 3(a)] that was approximately 34% narrower than that observed in the experiment [solid curve in Fig. 3(a)] [21]. To correct for this, the chirp rate was used as a fitting parameter. When it is reduced by 27%, the self-phase-modulated idler spectrum in the simulation [dotted curve in Fig. 3(a)] agrees well with that obtained by experiment [solid curve in Fig. 3(a)]. The simulated spectrum [dotted curve in Fig. 3(a) and experiment [solid curve in Fig. 3(a)] bandwidths differ by only 4%. Except for the chirp rate, the parameters used in the simulation were the same as the experimental parameters [21]. When the chirp rate was fixed at 500 fs^2 (pulse duration of 52 fs), the chirp rate of the

self-phase-modulated idler (after spectral broadening) in the simulation [dotted curve in Fig. 3(b)] agreed very well with that of the experiment [solid curve in Fig. 3(b)]. These results indicate that the reduced chirp rate (500 fs²) is close to that in the experiment.

The temporal shape of the idler pulse immediately after spectral broadening by SPM consists of a single peak; however, it changes drastically during subsequent propagation in the transparent medium due to GVD [Fig. 3(b)]. The upchirp in the center region of the idler is enhanced by the positive GDD, and the temporal intensity of the center region subsequently decreases. On the other hand, the down-chirps at the leading and trailing edges of the pulse are partially compensated by GVD and form intense spikes. The numerical simulation qualitatively agrees with the experimental intensity profile obtained by measuring second-harmonic frequencyresolved optical gating for the idler [Fig. 3(b)] [37]. The discrepancy in the relative peak heights obtained by the simulation and the experiment may be due to the different propagation modes in the fiber in the experiment and the simulation; in the experiment, the spatial beam profile may be modulated by self-focusing and subsequent photoionization in front of the fiber and during propagation inside the fiber [38,39], which might affect the nonlinear propagation process (i.e., the spectral broadening process) in the hollow fiber, whereas, in the simulation, the propagation mode inside the fiber is assumed to be the EH11 mode [36]. Spectral broadening by SPM may also be affected by the high-order spectral phase of the input pulse, which may be induced in a Ti:sapphire chirped-pulse amplifier that has several transparent media, a stretcher, and a compressor.

In the simulation, the generated signal pulse is negatively chirped and is compressed by propagating through a 0.9 mm thick plate of MgF_2 (the output window of the hollow-fiber chamber) and through 1.4 m of air [this length is optimized



Fig. 3. (Color online) Simulated (dotted curves; the chirp rate of the idler is 500 fs²) and experimental (solid curves) pulse shapes. Spectra of the (a) idler and (c) signal. (b) Temporal intensity profiles and instantaneous frequency change $-\Delta\omega$ with respect to time of the self-phase-modulated idler. (d) Temporal intensity profile of the compressed signal. In (a) and (c), the intensity-weighted average frequency of each spectrum is set to zero. The broken curve in (a) indicates the spectrum of the idler simulated using the estimated chirp rate.

to give the shortest compressed pulse duration; Fig. 3(d)]. The optimum length of air in the simulation (1.4 m) differs from that in the experiment (1.9 m) [21] due to the different temporal shapes of the idlers used for BCP-FWM in the simulation and the experiment. However, the spectrum of the calculated signal pulse agrees well with the experiment [Fig. 3(c)] [21]. This is because the instantaneous frequency and the temporal amplitude of the self-phase-modulated idler vary smoothly with time in the center region of the temporal profile in the simulation and the experiment. Additionally, the envelope of the pump pulse is nearly Gaussian, and the instantaneous frequency varies linearly with time in the simulation and the experiment. These characteristics do not change greatly when the parameters for the spectral broadening of the idler (such as the gas pressure and the pulse energies) are varied. Although the simulation and the experiment have different intensity profiles for the idler at the leading and trailing edges, they do not contribute greatly to signal generation due to the poor overlap between them and the pump pulse.

The signal spectrum contains a fine modulation structure with a modulation amplitude. This is because of the components with nonlinear frequency chirps at the leading and trailing edges of the idler [Fig. 3(b)] that have a small temporal overlap with the pump pulse. However, the modulation amplitude in the signal spectrum is smaller than that in the idler spectra, leading to a single-pulse shape for the transformlimited pulse, whereas the transform-limited pulse shape of the idler is not a single pulse. The signal pulse shapes after compression by normal GVD in the transparent media are single pulses in the simulation and the experiment [Fig. 3(d)]. The pulse widths of the compressed signal pulses are 9.7 and 8.2 fs, respectively, for the experiment and the simulation, and the transform-limited pulse widths estimated from the spectra in Fig. 3(c) are 7.8 and 7.7 fs, respectively; thus, there is good agreement between the experiment and the simulation.

A nearly Gaussian-shaped spectrum is obtained for the signal pulse when the pulse duration of the pump pulse is considerably shorter than that of the idler. An example of such signal generation can be simulated using a 35 fs Gaussian pulse (center wavelength, 800 nm; pulse energy, 1.5 mJ). The NIR pulse is chirped to 150 fs using a 51 mm thick fused-silica plate before it is coupled into a Kr-filled hollow fiber (core diameter, 140 μ m; length, 1 m; gas pressure, 2 atm). The selfphase-modulated idler from the fiber passes through the 1 mm thick MgF₂ output window of the hollow-fiber chamber, air of 2 m, and the 0.5 mm thick MgF_2 input window of the hollowfiber chamber for BCP-FWM. A 27 fs 400 nm pump pulse is generated by frequency doubling a 35 fs NIR pulse in a 100 μ m thick BBO crystal. The pump pulse is then chirped to 128 fs using a double-pass Brewster fused-silica prism compressor with an apex separation of 390 mm before it is combined with the broadband idler using a 0.5 mm thick dichroic mirror and focused into the second hollow fiber (for BCP-FWM; core diameter, 250 μ m; length, 0.6 m) filled with Ar gas of 0.03 atm. The pump and the idler coupled into the fiber are assumed to have pulse energies of 1 and 0.5 mJ, respectively.

Under these conditions, an idler pulse with a broad spectrum supporting a transform-limited pulse duration of 3.5 fs is concluded to be generated. The temporal width of the up-chirp component of the self-phase-modulated idler ex-

ceeds 200 fs, which is longer than the pulse duration of the pump pulse [128 fs; Fig. 4(a)]. The temporal intensity of the idler in this temporal region is flat (within 26%). Thus, the temporal shape of the signal is mainly determined by the pulse shape of the pump pulse (which is nearly Gaussian), as indicated by Eq. (1) and the calculated temporal intensities of the signal and pump shown in Figs. 4(a) and 4(c); this result is similar to those of the simulations and experiments described previously. The temporal intensity profile and linear frequency chirp of the signal pulse are realized using the interaction between the linearly chirped component of the self-phase-modulated idler and the linearly chirped pump pulse. The transform-limited pulse shape of the signal, which is calculated from the signal spectrum, has a single-pulse shape with a duration of 5.4 fs. Its transform-limited pulse duration is longer than that of the idler (3.5 fs) due to the idler having a longer pulse duration than the pump pulse; the frequency components in the center region (the up-chirp component) of the idler interact with the pump pulse to generate the signal, whereas the low- and high-frequency components at the leading and trailing edges of the idler do not contribute to signal generation. In other words, to realize an excellent Gaussian spectral shape, it is essential to consider that not all of the frequency components in the idler can be used for BCP-FWM and to ensure that the self-phase-modulated idler has a broader bandwidth than the signal.

By propagating the signal pulse through the 0.5 mm thick output MgF₂ window of the hollow-fiber chamber and then through 1.3 m of air, the negative frequency chirp in the signal is compensated for allowing a single compressed pulse to be generated [Fig. 4(d)]. The compressed profile contains no satellite pulses because the propagation length of the signal pulse in the transparent medium is not large enough to induce appreciable high-order spectral phase distortion. The signal pulse duration after dispersion compensation is 5.4 fs, which is almost the same as the transform-limited pulse duration (5.4 fs). Nearly transform-limited sub-10 fs UV pulses with excellent temporal and spectral profiles can therefore be obtained by BCP-FWM. When the negative GDD in the generated signal pulse exceeds that in the simulation, the propagation distance in air for compensation becomes longer. This longer propagation distance in air provides larger third-order dispersion, resulting in the generation of satellite pulses in the temporal profile of the compressed pulse. Consequently, the temporal shapes of the signal pulse diverge from a single-pulse shape because of the broad spectral width of the signal supporting the sub-6 fs transform-limited pulse duration.

A Gaussian-like signal spectrum has been obtained experimentally (Fig. 5), although the pulse energy of the Ti:sapphire laser was not sufficiently high to realize the above situation, and the obtained spectral widths of the idler and signal were narrower. When a 100 fs pump pulse and a more narrow-band self-phase-modulated idler than that used in the above experiment [21] are used, the signal pulse has the smooth spectral shape shown in Fig. 5(a), whereas the spectral shape of the idler pulse contains three spectral peaks. The compressed pulse shape in this case is also a single pulse, which is the same as in the experiment (Fig. 3; [21]) and the simulation (Figs. 3 and 4). The signal bandwidth is broader than that of the idler, which is opposite to the simulation results (Fig. 4). This is because the ratio between the temporal width



Fig. 4. (Color online) Simulated pulse shapes: (a) temporal intensity profile and instantaneous frequency change of the self-phase-modulated idler (solid curve) and temporal intensity profile of the pump pulse (broken curve). (b) Spectra of the idler (broken curve) and signal (solid curve). (c) Temporal intensity profile and instantaneous frequency change with respect to time. (d) Temporal intensity profile of the compressed signal (solid curve) and the inverse Fourier transform of the spectrum in (c). In (b), the carrier frequency corresponding to a wavelength of 800 nm is set to zero for the idler, while the third-harmonic wavelength of 800 nm (267 nm) is set to zero for the signal.

of the up-chirp component of the idler pulse to the pump-pulse duration in the experiment is smaller than that in the simulation. In the experiment, the pump pulse interacts with a large portion of the frequency components of the idler, which gives the signal a broader spectral width than the idler. In contrast, the pump pulse interacts with a limited portion of the frequency components of the idler in the simulation. In the experiment, the pump pulse interacts with the down-chirp portions of the idler to a certain degree so that the experimental signal spectrum deviates slightly from a pure Gaussian spectral shape. However, comparison of the experimental and simulation results demonstrates that BCP-FWM enables a single pulse to be generated with an almost single peak, almost Gaussian spectral shape [Figs. 4(b) and 5(a)].

In the previous discussion, the idler is chirped before the idler spectrum is broadened by SPM. Without such prechirping, it is difficult to generate a DUV pulse with excellent

temporal and spectral intensity profiles. A broadband idler supporting a 3.5 fs pulse can be generated by spectrally broadening a 35 fs transform-limited pulse with a pulse energy of 100 μ J through SPM in a Kr-filled (0.2 atm) hollow fiber. A positively chirped broadband idler can be obtained by broadening the pulse duration using transparent media [7 mm thick CaF_2 , 1.5 mm thick MgF₂, and 2 m of air; Figs. 6(a) and 6(b)]. The temporal width of the up-chirp component in the idler is limited, but the whole pulse duration is long, as shown in Fig. 6(a). Consequently, the BCP-FWM induced by the idler generates a signal pulse with complicated intensity and phase profiles [Fig. 6(c)] as well as a complex spectrum [Fig. 6(b)]. Although the signal pulse duration can be compressed by normal GVD when passing through 0.5 mm thick MgF₂ and 1.7 mof air, the compressed pulse is not a single-peaked pulse [Fig. 6(d)]. This is because the frequency chirp of the idler is not linear but contains a complicated structure within the temporal region of the interaction where the idler overlaps



Fig. 5. (Color online) Experimental pulse shapes: (a) spectra of the signal (solid curve) and self-phase-modulated idler (dotted curve). The intensity-weighted average frequency is set to zero in both curves. (b) Temporal intensity profile of the compressed signal.



Fig. 6. (Color online) Simulated pulse shapes: (a) Temporal intensity profile and instantaneous frequency change of the self-phase-modulated idler (solid curves) and temporal intensity profile of the pump pulse (broken curve). (b) Spectra of the idler (broken curve) and signal (solid curve). (c) Temporal intensity and instantaneous frequency change with respect to time. (d) Temporal intensity profile of the compressed signal (solid curve) and the inverse Fourier transform of the spectrum in (c) (dotted curve). In (b), the carrier frequency corresponding to a wavelength of 800 nm is set to zero for the idler, while the third-harmonic wavelength of 800 nm (267 nm) is set to zero for the signal.

with the pump pulse. Thus, prechirping the idler pulse prior to spectral broadening by SPM is essential to generate sub-10 fs DUV pulses with Gaussian spectral and temporal profiles (Fig. 4).

The following requirement must be considered in order to generate sub-10 fs single DUV pulses by BCP-FWM. Since the pump pulse has a narrow spectral width, the third-order dispersion in the pump pulse arising from the prism compressor does not induce substantial modulation of the intensity and phase profiles of the pump pulse and, thus, does not lead to large high-order phase distortion in the signal pulse. However, if the spectral width of the pump pulse is increased and the pump pulse is longer than 1 ps, the effect of third-order dispersion may be substantial and may generate subpulses in the compressed signal pulse. When generating 5 fs UV pulses, high-order dispersion during propagation of the signal pulse in air cannot be neglected for propagation distances of several meters, and third-order dispersion due to air creates subpulses in the compressed temporal profile of the signal pulse and the pulse duration may be broader than 5 fs: this is confirmed by a calculation that shows that the temporal profile of a 5 fs Gaussian pulse is modified by third-order dispersion in air so that it contains several postpulses. In addition, the propagation distance in a transparent medium with a large high-order GVD must be as short as possible after spectral broadening of the idler by SPM; otherwise, the temporal intensity and phase profiles will be distorted by high-order dispersion in air because of the broad bandwidth. The distortion in the idler may affect the temporal intensity and phase (and hence the spectral intensity and phase) of the signal pulse, and result in the compressed signal pulse deviating from smooth temporal and spectral profiles.

In real applications of this method that employ a chirped input pulse to generate a negatively chirped DUV output pulse, such as BCP-FWHM and the methods described in [15–17,21], the following consideration must be taken into account. A negative GDD in a device such as a prism compressor is associated with substantial spectral phase distortion due to third- and higher-order dispersions. This distorts the temporal profile and the phase of a laser pulse with a broad spectrum that has a Fourier-transform-limited pulse duration shorter than 10 fs. The distortion is transferred to the output DUV pulse. The pulse duration of the DUV pulse is determined by the distortion and is longer than 10 fs. Sub-10 fs DUV pulses could not be obtained by passing a broadband pulse through a prism compressor before or after frequency doubling [11,17]. A prism compressor is used in the method proposed in this paper, but it is used to negative chirp the narrow-band pump pulse. The temporal profile and phase of the pump pulse are not distorted much by the high-order spectral phase distortion induced by the prism pair. In addition, the optical path length of the self-phase modulated idler in transparent media is minimized. These conditions enabled sub-10 fs DUV pulses to be generated, as demonstrated in [21].

Four-wave mixing is a nonlinear optical process in which the signal generation efficiency is proportional to the idler intensity and the square of the pump intensity. Using a pump pulse (1 mJ) and a broadband idler (0.5 mJ) whose energies are more than 10 times higher than those in the experiment described in [21], the signal pulse energy will be 3 orders of magnitude higher than that in [21]. The DUV pulse energy calculated in the simulation exceeds 100 μ J. However, in practice, multiphoton ionization and self-focusing in front of the hollow fiber for SPM may degrade the beam profile of the output broadband idler from the hollow fiber. This may reduce the coupling efficiency of the broadband idler to the second hollow fiber (hollow fiber for FWM), and the pulse energy of the DUV pulse may be limited to several tens of microjoules in practical cases. Wojtkiewicz *et al.* performed an experiment in which a DUV pulse was generated with a pulse energy of 65 μ J by using a 1 mJ pump pulse in CP-FWM [16]. Sub-10 fs DUV pulses with pulse energies of several tens of microjoules are expected to be realized using the BCP-FWM technique discussed in this paper.

The maximum pulse energy of the signal pulse is also limited by the depletion of the pump pulse. A substantial degree of pump depletion will affect the temporal intensity and phase. Since a signal pulse energy of several tens of microjoules is less than 10% of the input pump-pulse energy (1 mJ), the effect of depletion is not considered to be large in this case. For higher-energy input pulses, it should be possible to keep this depletion small by employing a hollow fiber with a large core diameter. To generate high-energy few-cycle DUV pulses by BCP-FWM, it may be necessary to indirectly control the spectral phase of the signal pulse by using a spatial light modulator to precisely control the spectral phase of the idler [40]. Precise control of the spectral phase of the signal pulse can be realized by indirect control of the spectral phase since the phase of the signal pulse is linearly related to that of the idler [Eq. (1)] and nonlinear phase modulation (such as SPM) has a low efficiency in BCP-FWM. This has the potential to generate sub-5 fs DUV pulses with better spectral phase control than the technique presented in this paper.

Three nonlinear phenomena occur in the BCP-FWM technique developed in this study: FWM when generating the DUV pulse, second-harmonic generation when producing the NUV pump pulse, and SPM when spectral broadening the idler pulse. The FWM is linear with respect to the idler, whereas it is nonlinear with respect to the pump pulse. Consequently, FWM enhances the fluctuations in the NUV pump pulse. Fluctuations in the NUV pump are caused by fluctuations in the pulse energy of the NUV pump pulse generated through the second-harmonic generation process. Fluctuations in the pulse energy of the NUV pulse can be minimized by saturating second-harmonic generation or by stabilizing the pulse energy of the input pulse to the second-harmonic generation crystal [41]. The pulse energy and spectrum of the DUV pulse fluctuate when those of the self-phase-modulated idler fluctuate. The fluctuations in the self-phase-modulated idler may arise from self-focusing and photoionization of the noble gas at the entrance of the hollow fiber for SPM. These unwanted phenomena occur when the peak intensity of the input idler pulse and/or the gas pressure at the entrance of the hollow fiber for SPM are high; they do not occur much at the entrance of the hollow fiber for FWM because of the low gas pressure there. Positively chirping the idler pulse (prechirping) prior to spectral broadening by the SPM reduces the peak intensity of the idler pulse and suppresses self-focusing and photoionization [42]. The stability of the idler pulse can be further improved by employing a pressure gradient [38,39,42]. Although the spatial mode profile of the spectrally broadened idler pulse may be degraded by self-focusing and photoionization, it does not affect the spatial mode profile of the DUV laser pulse greatly since the beam profile of the DUV pulse is determined by the propagation mode in the hollow fiber, in which the gas pressure is low. Using a beam-pointing stabilizer [6,43,44] imparts high stability to the DUV pulse. This technique has been used to generate a 10 fs NUV laser pulse that is sufficiently stable to be used for spectroscopy even though two nonlinear optical phenomena (second-harmonic generation and SPM) are involved [6]. Using beam-pointing stabilizers for the input idler and pump pulses would also result in a stable spatial beam profile for the DUV pulse.

The technique of BCP-FWM is also applicable to the generation of ultrashort pulses in the vacuum UV (VUV) as well as in the DUV region. For generating a VUV pulse, the 400 nm (NUV) pump pulse is replaced by a 267 nm (DUV) pulse that is the third harmonic of the output of a Ti:sapphire chirpedpulse amplifier. For negatively chirping the third-harmonic pump pulse, a prism pair made of calcium fluoride or magnesium fluoride should be used rather than that made of fused silica [11]. By use of the chirped third-harmonic pump pulse and a self-phase-modulated idler (center wavelength of 800 or 400 nm), a sub-10 fs VUV pulse is expected to be generated in the VUV region (center wavelength of 160 or 200 nm).

The shortest pulse duration obtained by BCP-FWM is around 5 fs, and is determined by the bandwidths of the input pulses. For generating shorter pulses, the input pump pulse should also be broadband. Spectral broadening of the pump pulse by SPM and subsequent negative chirping of it are necessary. A pulse stretcher for negative chirping, such as a prism pair or chirped mirrors, however, induces high-order dispersion, and may distort the temporal shape of the broadband pump pulse. This is expected to affect the temporal profile and phase of a signal generated by BCP-FWM, and may make it difficult to generate sub-5 fs pulses after the dispersion compensation. This might be necessary to be considered for generating sub-5 fs DUV pulses by BCP-FWM.

5. CONCLUSION

Sub-10 fs DUV pulses are generated by BCP-FWM, which produces single DUV pulses without using a pulse compressor (except for a prism compressor for the relatively narrow-band input pump pulse). Prechirping the idler is critical when generating single pulses, since it expands the temporal width of the linearly chirped pulse in the self-phase-modulated idler. This technique does not require precise spectral phase control by using a spatial light modulator, a deformable mirror, and/or a dazzler; rather, only a prism compressor is used to negatively chirp a relatively narrow-band pump pulse. Since the pump has a narrow bandwidth, the prism compressor may be replaced with chirped mirrors for NUV pulses. Although chirped mirrors have oscillations in their GDD curves, these oscillations do not greatly distort the temporal shape of the pump pulse because of the narrow bandwidth. Replacing the prism compressor with chirped mirrors should make BCP-FWM more robust than the setup used in the experiment described in [21]. Since this method uses chirped (low-intensity) pulses for both spectral broadening of the idler by SPM and frequency conversion by BCP-FWM, using high-energy laser pulses will not cause appreciable photoionization. Thus, this method is not limited to generating low-energy sub-10 fs pulses, but it can also be used to generate high-energy sub-10 fs pulses that are useful for investigating high-order harmonic generation and ultrafast photoelectron spectroscopy in the DUV region. This technique generates sub-10 fs DUV pulses with smooth shapes that are close to Gaussian temporal and spectral profiles that are ideal for spectroscopy. The demonstration of such high-energy sub-10 fs DUV laser systems is expected to open up new research areas in ultrafast physics and chemistry in the DUV wavelength region.

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