Generation of 2.6 fs optical pulses using induced-phase modulation in a gas-filled hollow fiber

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Received November 16, 2006; accepted December 8, 2006;
posted January 2, 2007 (Doc. ID 77116); published March 15, 2007

We report quasi-one-optical-cycle pulse compression of the ultrabroadband white-light continuum generated using both induced-phase modulation (IPM) and self-phase modulation (SPM) in a 3.0 atom Ar-gas-filled hollow fiber. Fundamental and second harmonic waves of amplified 30 fs Ti:sapphire laser pulses were irradiated into a 37 cm hollow fiber with an inner diameter of 140 μm. When the two pulses were temporally overlapped in the hollow fiber, the white-light continuum with the wavelength range of 350–1050 nm was generated. The spectral phase of the white-light continuum was measured by a modified spectral interferometry for direct electric-field reconstruction, and quasi-automatic feedback chirp compensation was carried out using a programmable liquid-crystal spatial light modulator placed on the Fourier plane of a 4-*f* system. As a result, 2.6 fs, 3.6 μJ, 1.3 cycle transform-limited (TL) pulses with a peak power up to 1.4 GW at a 1 kHz repetition rate were generated in the visible to near-infrared region (the over-one-octave bandwidth of 450–975 nm). The fact that the IPM+SPM light was compressed to the TL duration is important toward the generation of a single, intense one-optical-cycle pulse in the visible region. © 2007 Optical Society of America

OCIS codes: 320.5520, 190.4370.

1. INTRODUCTION

Generation of intense, few-optical-cycle pulses has been attracting a lot of interest especially for their applications to high harmonic generation.[1] In many studies, white-light continuua generated in noble-gas-filled hollow fibers (HFs),[2] or using filaments in noble gases[3] have been compressed with chirp mirrors[4] or spatial light modulators (SLMs).[5] Our group has been adopting the Ar-gas-filled HF and the programmable liquid-crystal SLM scheme toward the generation of one-optical-cycle pulses in the visible region. As a result, the generation of 2.8 fs, 0.5 μJ pulses has already been achieved.[6] In the study, the white-light continuum was generated by only self-phase modulation (SPM), which limits the bandwidth and the output pulse energy. So, another method has been needed to generate optical pulses with shorter duration and higher energy.

Induced phase modulation (IPM)[7,8] is a prominent phenomenon to generate broader-band optical pulses than those produced by only SPM. It is based on the interaction of copropagating two (or more) different-colored optical pulses, and has a controllability of the spectral structure by adjusting the intensity ratio and relative time delay of input pulses. We have already reported the generation of ultrabroadband optical pulses using the IPM effect not only in glass fibers,[9] but also in Ar-gas-filled HFs.[10] Using a glass fiber, the 530–880 nm continuum has been compressed to 7.8 fs while the transform-limited (TL) duration for the spectrum was 4.4 fs.[11] This difference seems to come from low-output pulse energy and strong dispersion of the glass fiber. Noble-gas-filled HFs have the advantages of both high damage threshold and low dispersion, so we have tried pulse compression using IPM as well as SPM in an Ar-gas-filled HF. As a result, the generation of 2.6 fs, 3.6 μJ, 1.3 cycle TL pulses with a wavelength range from 450 to 975 nm, a center wavelength of 600 nm and a peak intensity of approximately 1.4 GW was achieved.

2. EXPERIMENT AND RESULT

The experimental setup is shown in Fig. 1. The output beam of a multipass Ti:sapphire laser amplification system (center wavelength, 800 nm; duration, 30 fs; repetition rate, 1 kHz; pulse energy, 1.3 mJ) was divided into two beams by a beam splitter (BS1) with a split ratio of 9:1 (=reflectance:transmittance). The weak pulse transmitted through BS1 served as a reference pulse for the modified spectral interferometry for direct electric-field reconstruction (M-SPIDER)[12] measurements. The intense pulse reflected by BS1 was divided by another beam splitter (BS2) with a split ratio of 1:2. The reflected pulse was passed through a 0.5 mm thick β-barium borate (BBO) crystal (type I). The second-harmonic (2ω) pulse generated under the phase-matching condition was guided to a pair of roof mirrors on a translational stage. The fundamental pulse transmitted through BS2, which served as the ω pulse, was introduced to periscopes (PS) so that its polarization was turned by 90°. Then the ω and 2ω pulses having the same polarization were temporally and spatially recombined by a dichroic mirror (DM) and focused into a fused-silica HF (length: 37 cm; inner diameter:

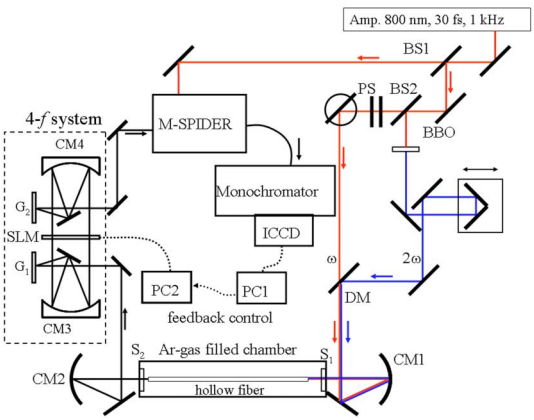

Fig. 1. (Color online) Experimental setup for extremely short-pulse generation. BS1, beam splitter with a split ratio of 9:1 (=reflectance:transmittance) BS2, beam splitter with a split ratio of 1:2. BBO, type I β-barium borate crystal with a 0.5 mm thickness. PS, periscopes. DM, dichroic mirror. CM1 and CM2, concave mirrors with a focal length of 400 and 200 mm, respectively. G_1 and G_2, gratings with a groove of 384.6 lines/mm. CM3 and CM4, concave mirrors with a focal length of 200 mm. ICCD, intensified CCD detector.

140 μm) by a concave mirror (CM1) with a focal length of 400 mm. The HF was placed in a chamber filled with 3.0 atm Ar gas. The chamber had two 1 mm thick fused-silica windows (S_1 and S_2). The white-light-continuum beam from the HF was collimated by a concave mirror (CM2) and guided to a 4-f system with an SLM. The 4-f system was composed of two aluminum concave mirrors with a focal length $f=200$ mm (CM3, CM4) and two silver-coated reflective gratings (G_1 and G_2) with a groove density of 384.6 lines/mm and with a blaze wavelength of 500 nm. The SLM consists of 648 pixels with a pixel width of 97 μm and a gray-scale resolution of 192.

The ω pulse with 182 μJ energy and the 2ω pulse with 116 μJ energy were focused and injected into the HF. The energies were chosen such that the IPM+SPM spectrum became the broadest and strong ionization of Ar gas did not occur. Output energy of the ω (2ω) pulse from the HF was 56 (28) μJ in the case of the one beam transmission, corresponding to the transmission efficiency of 31% (24%). When the two pulses were temporally overlapped at the exit of the HF to enhance the IPM effect,[10] the total output pulse energy with the spectral broadening from 350 to 1050 nm was 67 μJ. A black curve in Fig. 2 shows the spectrum employed for the pulse compression; both the edges were clipped by a slit on the Fourier plane in the 4-f system owing to technical reasons described later. Gray curves show the spectra observed when the two pulses propagated separately in the HF. In this case, the spectral broadenings are caused by only the SPM effect. Compared to the SPM spectrum (by the ω pulse), the spectral component of the IPM+SPM light with wavelengths shorter than 550 nm is enhanced owing to the IPM effect. As a result, the white-light continuum with an almost flat spectrum over the whole wavelength range except for the 750–850 nm range was obtained.

Only the single-mode component of the output beam from the HF was clipped by an iris with an aperture of approximately 3 mm. After that, the ultrabroadband pulse with an energy of 26 μJ was guided to the 4-f chirp compensator. The output pulse with the energy of 3.6 μJ was guided to the M-SPIDER system, where two replica pulses with a relative delay of $\tau=856$ fs were generated by a Michelson interferometer. The replica pulses were upconverted by the external chirped pulse in a 20 μm thick BBO crystal (type II). The external chirped pulse was originally divided from the output of the Ti:sapphire laser amplifier and was passed through a 10 cm long TF5 glass twice for chirping. The replica pulses and the chirped pulse were focused onto the BBO crystal by a 50 mm focal-length parabolic mirror with a crossing angle of ~0.17 rad, and the M-SPIDER signal was generated in the middle direction. A calcite prism polarizer was placed to cut the fundamental light and detect only the M-SPIDER signal. The interferogram with a spectral shear $\Omega/2\pi=4.11$ THz was observed using a spectrometer with an intensified CCD array detector, as shown in Fig. 3. The signal ranges from 290 to 445 nm without any discontinuity. The retrieved spectral phase before chirp compensation is drawn by a broken curve in Fig. 4, and it ranges over 300 rad in the whole spectral range. Here, we note the bandwidth of optical pulses measurable by using our M-SPIDER system. In general, an over-one-octave

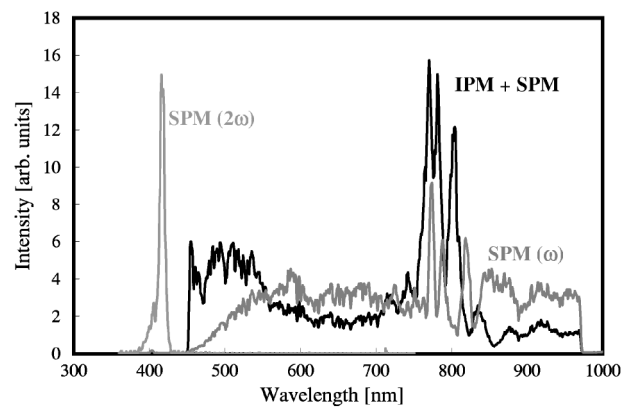

Fig. 2. Spectra of ultrabroadband optical pulses generated by only SPM effect (gray curves) and by SPMs and IPM (black curve). Input pulse energy of $\omega(2\omega)$ pulse was 182 (116) μJ. Output pulse energy of the IPM+SPM light was 67 μJ.

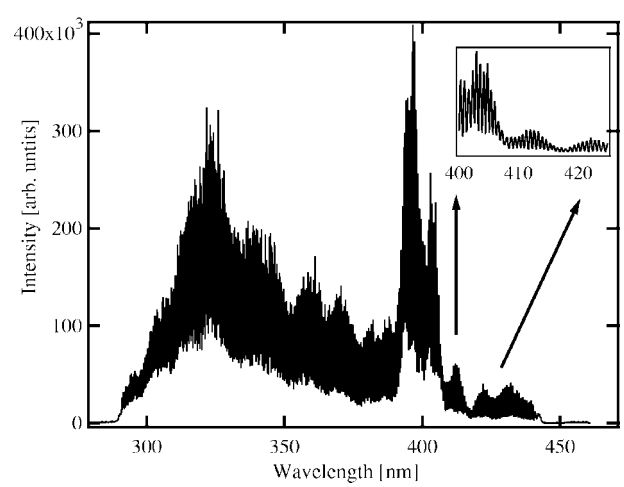

Fig. 3. M-SPIDER signal before feedback chirp compensation.

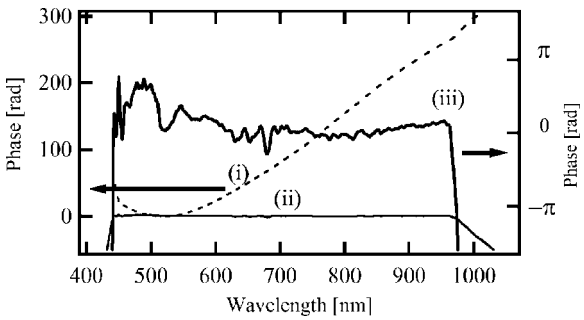

Fig. 4. Spectral phases (i) before (broken curve) and (ii) after (thin solid curve) second feedback chirp compensation (scale is left axis). (iii) Expanded spectral phase of the second feedback (thick solid curve, scale is right axis).

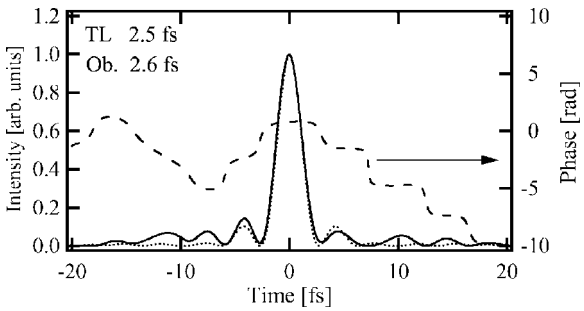

Fig. 5. Temporal intensity profile of compressed pulses (solid curve) and that of TL pulses (dotted curve). Broken curve shows the temporal-phase profile of compressed pulses.

pulse cannot be measured accurately owing to the second-order diffraction from the gratings used in a 4-*f* system. However, by appropriately choosing the blaze wavelength of the gratings, the intensity of the second-order diffraction can be reduced a lot; if it is chosen to be 500 nm as in the present study, the intensity of second-order diffraction is only a few percent of the first-order one. Therefore, the spectral-phase measurement of the over-one-octave pulse has already been possible.[13] After the second feedback chirp compensation using the M-SPIDER signal and the SLM, the spectral phase became almost flat over the one octave frequency range; the phase variation was within π rad as shown in Fig. 4. Figure 5 shows the retrieved temporal profile of the pulse after the second feedback compensation (solid curve) and that of the TL one (dotted curve) for the IPM+SPM spectrum shown in Fig. 2. The duration of the compressed pulse and that of the TL one are 2.6 fs and 2.5 fs, respectively, corresponding to 1.3 cycles with a center wavelength of 600 nm. The pulse energy was 3.6 μJ so that the peak power was 1.4 GW at a repetition rate of 1 kHz. The time-dependent phase drawn as a broken curve in Fig. 5 is also flat during the pulse duration. To the best of our knowledge, this is the shortest single pulse in the visible to near-infrared region.

3. DISCUSSION

Now we discuss our experimental results. First, we discuss the reason that the energy of compressed pulses has been increased up to 3.6 μJ compared with that (=0.5 μJ) of the previous study.[6] One reason is the use of the IPM effect. In the present study, input energies of the ω and 2ω pulses to the HF are 182 and 116 μJ, respectively, and the output pulse energy from the HF is 67 μJ, while in the previous study, only the ω pulse with an energy of 135 μJ was incident and the output energy was 18 μJ. The input pulse energy to an HF is limited by the ionization threshold of Ar gas; if it occurs, the single-mode output beam cannot be obtained and the spectrum of the output pulse becomes strongly modulated so that it is difficult to compress it. In the present study, two-colored input pulses with center wavelengths of 800 and 400 nm are incident to the HF, and they propagate with different group velocities and overlap at the exit of the HF to enhance the IPM effect.[10] By the time they overlap there, the peak intensities of them with respective center wavelengths have been much decreased, because large parts of the energies have been frequency-shifted through the IPM and SPM effects. As a result, the pulses with a higher energy in total than the ionization threshold in the case of the one-colored pulse can be incident without causing ionization of Ar gas. Thus the IPM effect is very useful to generate intense and broadband optical pulses. Another reason is the use of the HF with a larger inner diameter of 140 μm; in the previous studies, an HF with 100 μm inner diameter was used. It has improved the transmission efficiency from 13% to 22%. Actually, the larger the inner diameter of an HF becomes, the higher pulse energy we need to obtain an HF-output pulse with enough spectral broadening. The high-powered Ti:sapphire amplification system with a maximum output pulse energy of 2.5 mJ has made it possible to cause enough spectral broadening through the IPM and SPM effects in the HF with 140 μm inner diameter.

Second, we explain the reason that the bandwidth of the pulse to be compressed (white-light continuum) was restricted from 450 to 975 nm. This was to avoid the influence of the carrier-envelope-phase (CEP) fluctuation of HF input laser pulses, and to avoid the spectral mixing of the short-wavelength-edge component of the white-light continuum with the long-wavelength-edge component of the M-SPIDER signal. Let us express the IPM+SPM spectrum using nonlinear propagation equations. We assume that the ω and 2ω pulses are copropagating in the HF that is parallel to the z direction. For each pulse j ($j=\omega,2\omega$), the electric field (linearly polarized in the x direction) can be written as $\mathbf{E}_j(\mathbf{r}_j,t)=\frac{1}{2}\hat{\mathbf{x}}\{F_j(x,y)A_j(z,t)\times\exp[i(\beta_{0j}z-\omega_j t+\phi_{0j})]+\text{c.c.}\}$, where β_{0j}, ω_j and ϕ_{0j} are a wavenumber of the propagation mode, a center angular frequency, and an initial constant phase for a pulse j. $F_j(x,y)$ represents the transverse distribution of the mode and $A_j(z,t)$ represents the slowly varying envelope part of the field. For convenience, we define two time coordinates that move with the group velocity of each pulse as $T_j=t-\beta_{1j}z+t_{0j}$, where β_{1j} is the inverse of the group velocity of the pulse j and t_{0j} is the initial time of the j pulse (for $j=\omega$, $t_{0j}=0$). Under the slowly varying envelope approximation, neglecting the very low dispersion of Ar gas, $A_j(z,T_j)$ satisfies[14]

$$\frac{\partial A_\omega}{\partial z}=-\frac{\alpha_\omega}{2}A_\omega+i\frac{n_2\omega}{c}[f_{\omega\omega}|A_\omega|^2+2f_{\omega 2\omega}|A_{2\omega}|^2]A_\omega, \quad (1)$$

$$\frac{\partial A_{2\omega}}{\partial z} = -\frac{\alpha_{2\omega}}{2}A_{2\omega} + i\frac{2n_2\omega}{c}[f_{2\omega 2\omega}|A_\omega|^2 + 2f_{2\omega\omega}|A_\omega|^2]A_{2\omega}, \tag{2}$$

where α_j represents a loss, n_2 is a nonlinear refractive index, and f_{jk} is a mode overlap integral between transverse modes of the j and k pulses. Equation (1) represents the spectral broadening of the ω pulse owing to both the ω and 2ω pulses, and Eq. (2) represents the spectral broadening of the 2ω pulse. The IPM+SPM spectrum is the absolute square of the sum of the above two electric-field components, and an effective discontinuity in group-delay has been known to exist between them.[11,15] Under the present experimental condition, in which the energy of the ω pulse is approximately 1.6 times higher than that of the 2ω pulse, the discontinuity is observed at rather short wavelength of 452 nm so that the white-light continuum employed for pulse compression mainly consists of the former component. Thus the discontinuity point was avoided as a first step to compress the IPM+SPM light. In this case, it has been confirmed that the variance of CEP does not influence the intensity spectrum of HF output pulses.[8] Therefore the IPM+SPM light was compressed to almost the TL pulse duration without CEP locking of amplified laser pulses.

Third, we explain another reason that the bandwidth of the pulse to be compressed (white-light continuum) was restricted from 450 to 975 nm. Since the wavelength of the M-SPIDER signal for the fundamental light at 975 nm corresponds to 444 nm, the spectral component of the white-light continuum with the wavelength range shorter than 444 nm disturbs the M-SPIDER signal for the wavelength range of the fundamental light slightly shorter than 975 nm. Although the polarization of the white-light continuum and that of the M-SPIDER signal should differ by 90° in principle, and the two beam spots were separated spatially as previously described, the white-light continuum was not completely cut by a calcite polarizer placed after the BBO crystal and then was slightly mixed with the M-SPIDER signal. This is because the signal intensity of M-SPIDER is much weaker than that of the white-light continuum and the polarization of the white-light continuum becomes slightly elliptic after it passes through the HF and the SLM made of a liquid crystal, which shows birefringence. One of the possible solutions to overcome this problem is the use of a polarizer after the 4-*f* chirp compensator. However, it has the disadvantages of reducing the output pulse energy and adding extra chirp to the white-light continuum. Another one is the use of a different-colored reference pulse, which is in progress.

Finally, we mention the significance of the present result. First, the duration of 2.6 fs is the shortest as a duration of the single pulse (not "a train of pulses"[16]) with a repetition rate of kilohertz order in the visible to near-infrared region, covering over-one-octave bandwidth, reported to date. Second, the fact that the IPM light generated in an HF can be compressed to a TL pulse is very important toward the generation of 1.5 fs, one-optical-cycle pulses in the visible region, because the generation of intense ultrabroadband optical pulses with a wavelength range from 350 to 1050 nm has already been possible. Third, the peak power of compressed pulses is 1.4 GW, and it gives a possibility of photon density up to 10^{14} W/cm^2, which is a key value for the high harmonic generations.[17]

4. CONCLUSION

In conclusion, we have generated 2.6 fs, 3.6 μJ, 1.3 cycle transform-limited optical pulses by compensating for the chirp of ultrabroadband light generated by both the IPM and SPM effects in an Ar-gas-filled hollow fiber. By overcoming some technical problems, it will be possible to generate intense one-optical-cycle pulses with a repetition rate of 1 kHz, which should serve as an optical source for ultrafast spectroscopy and attosecond science and technology.[17]

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