Simultaneous compression and amplification of a laser pulse in a glass plate

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Abstract: We demonstrated a novel method of simultaneous compression and amplification of a weak laser pulse in a glass plate using cross-phase modulation in conjunction with four-wave optical parametric amplification that was pumped by an intense femtosecond pulse. A proof-of-principle experiment succeeded in smooth broadening of the weak pulse spectrum by a factor of about three and simultaneously amplifying the pulse energy by more than three times. By using chirped mirrors to compensate the dispersion, the weak pulse was compressed from 22.6 fs to 12.6 fs. Furthermore, the output spectrum of seed pulse could be tuned by varying the delay of the intense pump pulse with respect to the weak seed pulse. This method can also be used for simultaneous spectral broadening of several weak beams with different wavelength at the same time.

OCIS codes: (190.4380) Nonlinear optics, four-wave-mixing; (190. 4410) Nonlinear optics, parametric processes; (190.7110) Ultrafast nonlinear optics; (320.5520) Pulse compression.

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1. Introduction

Over the past decade, several pulse-compression techniques based on phase modulation induced by nonlinear optical effects have been developed extensively. One of the important methods to obtain compressed pulse is using self-phase modulation (SPM) in a gas-filled hollow fiber [1,2], a filament in gas cell [3,4], a fiber [5], or a bulk medium [6,7] to broaden spectrum and then compensating the spectral phase dispersion. This method is usually used to generate intense few-cycle pulses. Another technique for compressing pulses that has been developed in recent years is molecular phase modulation (MPM) [8,9]. In this method, a short, intense pump pulse excites impulsively coherent vibration in a Raman-active molecular gas, and then a weak, delayed pulse is phase modulated by the instantaneous refractive index change associated with the molecular vibration. A third pulse compression method is cross-phase modulation (XPM). It was extensively studied more than twenty years ago in a long fiber and in thick glass using picosecond pulses [10–13]. Recently, it has been theoretically demonstrated that XPM can compress a pulse to nearly the single-cycle regime from ultraviolet (UV) to mid-infrared (MIR) in a bulk medium [14]. Prior to the SPM effect, in the XPM process, the spectral, temporal, and spatial properties of the weak seed pulse can be controlled through manipulating both the intense pump pulse profile and the delay time between pump pulse and seed pulse [10,12,13]. Compared to MPM, it is much simpler and flexibility to broaden the spectrum using XPM in a bulk medium. In addition, the pump and seed beams are spatially well separated and the spectral shape is not limited by the time period of the molecular vibration [8,9].

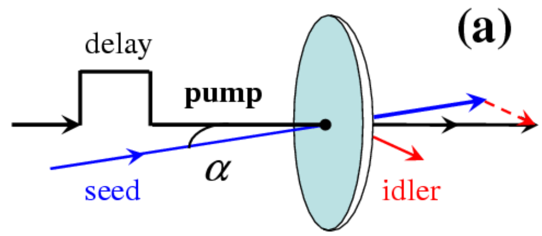
In this paper, we proposed and demonstrated for the first time a method for simultaneous compression and amplification of a weak femtosecond pulse in a bulk medium using XPM in conjunction with the four-wave optical parametric amplifier (FWOPA) [10,15–17] that is pumped by an intense femtosecond pulse. Furthermore, the spectrum of the weak pulse can be tuned by varying the delay between the pump and seed pulses. This method promises to be useful for the generation and optimization of ultrashort pulses at different wavelengths for use in pump–probe experiments that require tunable short pulses over a wide spectral range.

2. Principle

The principle of this method is schematically illustrated in Fig. 1(a). An intense pump beam and a weak seed beam are focused onto a glass plate with a crossing angle α. Usually, the wavelength of the pump pulse is fixed; it should be different from that of the seed pulse to prevent interference between the two pulses. When the pump and seed pulses are synchronous in time and overlapping in space in a transparent bulk medium, the seed pulse spectrum will be broadened due to the XPM effect in the medium induced by the intense pump pulse. Furthermore, the weak seed pulse will be simultaneously amplified when the crossing angle α satisfies the phase-matching condition of four-wave mixing (FWM) [10,15–17].

#117786 - $15.00 USD Received 28 Sep 2009; revised 9 Dec 2009; accepted 10 Dec 2009; published 22 Jan 2010

A diagram of the vectors of FWM is also presented in Fig. 1(a). According to the phase-matching condition, the crossing angle α_{in} in the medium can be represented by $\cos\alpha_{in} = \left[(2k_p)^2 + k_s{}^2 - k_i{}^2\right]/4k_p k_s$, in terms of wavenumber k with the subscripts of p, s, i indicating the pump, seed, and idler beams, respectively. Figure 1(b) and its inset show the plots of the phase-matching curves of the crossing angle in air α as a function of the seed wavelength for fused silica and CaF_2, respectively. The pump pulse is fixed at a typical wavelength of 800 nm and 400 nm for fused silica and CaF_2, respectively. As indicated in Fig. 1(b) [or inset of Fig. 1(b)], there is a broad phase-matching bandwidth around 500 nm (or 250nm) when the crossing angle α is about 3.1° (or 6.4°).



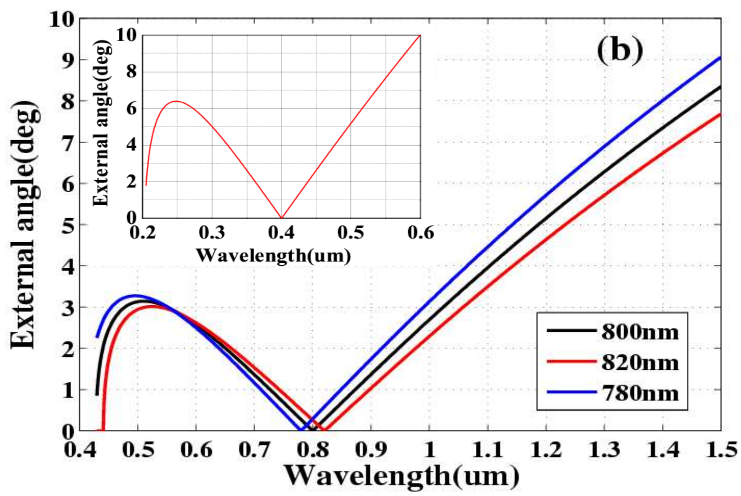


Fig. 1. (Color online) (a) Schematic of the experimental setup; α is the crossing angle. (b) Phase-matching curves showing the crossing angle α as a function of the seed wavelength for fused silica (or CaF2, inset) when the pump pulse was fixed at a typical wavelength of 780, 800, and 820 nm (or 400 nm, inset).

3. Experimental setup

The experiment was performed using a 1-kHz Ti:sapphire regenerative amplifier laser system (Micra + Legend-USP, Coherent) with a pulse duration of 40 fs and an average power of 2.5 W. Figure 2 showed experimental setup. The laser pulse was split into four beams. One of the beams (Beam3) was focused by a 1-m focal length lens onto a 0.5-mm-thick fused silica plate (G2) after passing through a variable neutral-density filter and a motor-driven delay stage with a resolution of 10-nm/step. Another beam (Beam4) was used to measure the pulse duration by the cross-correlation frequency resolved optical gating (XFROG) technique by mixing it with the two input beams or the spectrally broadened pulse in a 10-µm-thick BBO crystal. The other two beams (Beam1 and Beam2) were used to generate cascaded FWM sidebands in a similar manner as in our previous studies [18–21] in which nearly transform-limited pulses were obtained when the input pulses were appropriately chirped. These cascaded FWM sidebands were used as incident seed beams and were focused onto the glass plate by a concave mirror with a focal length of 600 cm. The diameter of the pump (seed) beam on the glass plate was 300 μm (153 μm) as measured using a CCD camera (BeamStar FX33, Ophir Optronics). The pulse duration of the pump (seed) pulse was about 75 ± 3 fs (22.6 ± 0.5 fs) prior to the glass. The group velocity delay between pump pulse (800nm) and seed (510nm/620nm) pulse was 36fs/14fs in a 0.5mm fused silica glass plate. The pump beam was focused into a larger diameter and its pulse duration was adjusted to be longer than that of the seed pulse to reduce the spatial chirp in XPM and ensure a good temporal overlap with the seed pulse through the glass plate.

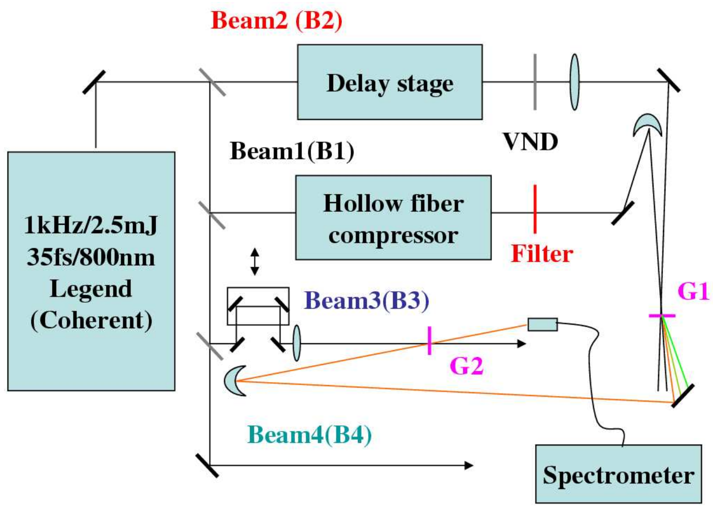


Fig. 2. (Color online) Experimental setup. VND: variable neutral-density filter. Filter: bandpass filter at 700nm center wavelength with 40 nm bandwidth. G1:1mm-thick CaF_2 glass plate. G2: 0.5-mm-thick fused silica glass plate.

4. Experimental results and discussion

Initially, the third-order anti-Stokes (AS3) sideband [18–21] was used as the seed beam because its center wavelength is close to the broad phase-matching spectral range around 500 nm. The incident pulse energies of the AS3 and pump beams were 300 nJ and 140 μJ, respectively. The spectral profile and spectral intensity of the output seed beam as a function of the delay time t_{ps} when the crossing angle α was 1.80° ± 0.05° are shown in Fig. 3(a). A negative delay time t_{ps} indicates that the seed pulse precedes the pump pulse. The full width at half maximum (FWHM) spectral bandwidth of the seed pulse was smoothly broadened from 18 to 50 nm (i.e., by a factor of about 2.8) at a delay time of approximately 0 fs due to XPM. This broadened spectrum can support transform-limited pulse duration of 9.6 fs. In this case, the spectral broadening of a Gaussian pulse due to XPM can be simply expressed as $\Delta\omega_s \approx \omega_s / cn_2(|E_s|^2 + 2|E_p|^2) l / T_0$ [10,13] because the intensity of the pump pulse is nearly undepleted and the group-velocity dispersion in the bulk medium is negligibly small. In the expression, ω_s is the angle frequency of the seed pulse, n_2 is the nonlinear refractive index of the medium, E_s and E_p are electric field amplitudes of seed pulse and pump pulse, respectively, l is the thickness of bulk medium, T_0 is the FWHM pulse duration of the pump pulse. Using the above expression, the spectral broadening is calculated to be about 40 nm in agreement with the experimental result. In this case, the seed beam was not amplified due to the large phase mismatch. When the crossing angle α was 3.30° ± 0.05°, the FWM phase-matching condition was satisfied [Fig. 1(b)]. In this case, the spectrum of the seed pulse was

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smoothly broadened and its output power was simultaneously amplified, as shown in Fig. 3(b). The pulse energy of the seed pulse was amplified from 300 to 940 nJ (i.e., by a factor of about 3.1) at a delay time of approximately 0 fs. The spectrally asymmetric amplification in the Fig. 3(b) is due to the fact phase matching is satisfied on the longer wavelength side for broader spectral range, as shown in Fig. 3(c). The phase-matching angle in the experiment (3.30° ± 0.05°) was slightly larger than the calculated value for the best phase matching as shown in Fig. 3(c) and Fig. 1(b).

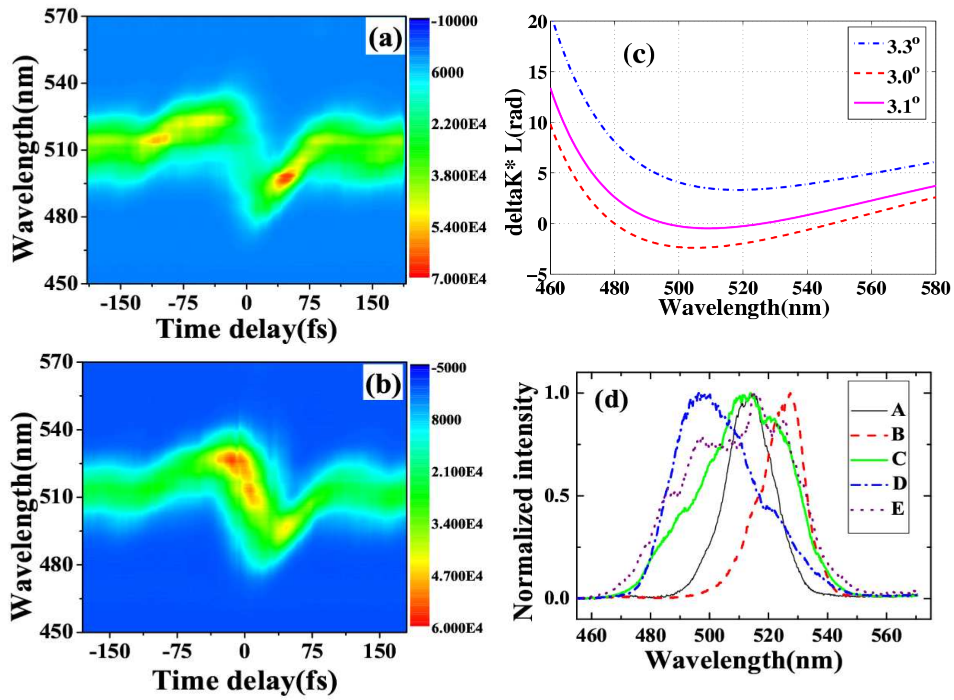

Fig. 3. (Color online) Dependence of the spectral profile and intensity of the output seed beam (AS1) on the delay time t_{ps} for crossing angles α of (a) 1.80° ± 0.05° and (b) 3.30° ± 0.05°. (c) The phase mismatching curves at three different external crossing angles (3.3°, 3.1°, and 3.0°) in a 0.5mm-thick fused silica glass when the seed pulse was centered at 510 nm and pumped by 800 nm pulse. (d) A: incident spectrum of seed pulse. B, C, and D are spectra of output pulse at delay times of −30, 0, and 30 fs when α was 3.30° ± 0.05°, respectively. E: spectra of output pulse at delay times of 0 fs when α was 1.80° ± 0.05°.

This is because of the nonlinear phase shift $\varphi_{NL} = \Delta k' l = 2\omega_p n_2 I_p l / c$ induced by the nonlinear index [10]. In the expression, ω_p and I_p are the angle frequency and intensity of pump pulse, respectively. This induced additional phase amounts to be about 3π phase in a 0.5 mm fused silica glass under the experimental condition. The spectra of the output seed pulse at delay times of −30, 0, and 30 fs are shown in Fig. 3(d). The seed pulse spectrum was clearly red-shifted (blue-shifted) when the two incident pulses were overlapping in negative (positive) delay time. The peak wavelength of the seed pulse can be shifted by about 20 nm on both sides, indicating that the spectrum of the amplified output pulse can be tuned by adjusting the delay time t_{ps}. This spectral shift can be easily explained as follows. The frequency shift induced by XPM can be given by $\delta\omega(t) = -\partial\varphi_{NL} / \partial t \propto -\partial \left|E_p(t)\right|^2 / \partial t$, where $E_p(t)$ is the electric field of the pump pulse. It can be concluded from the above expression that $\delta\omega(t) < 0$ at the leading edge of the pump pulse and $\delta\omega(t) > 0$ at the trailing edge of the pump pulse. As for the negative delay time, the leading edge of the pump pulse will overlap

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with the seed pulse. Therefore, the induced frequency change is negative ($\delta\omega(t) < 0$) and the spectrum of the seed pulse is red-shifted; vice versa for a positive delay time.

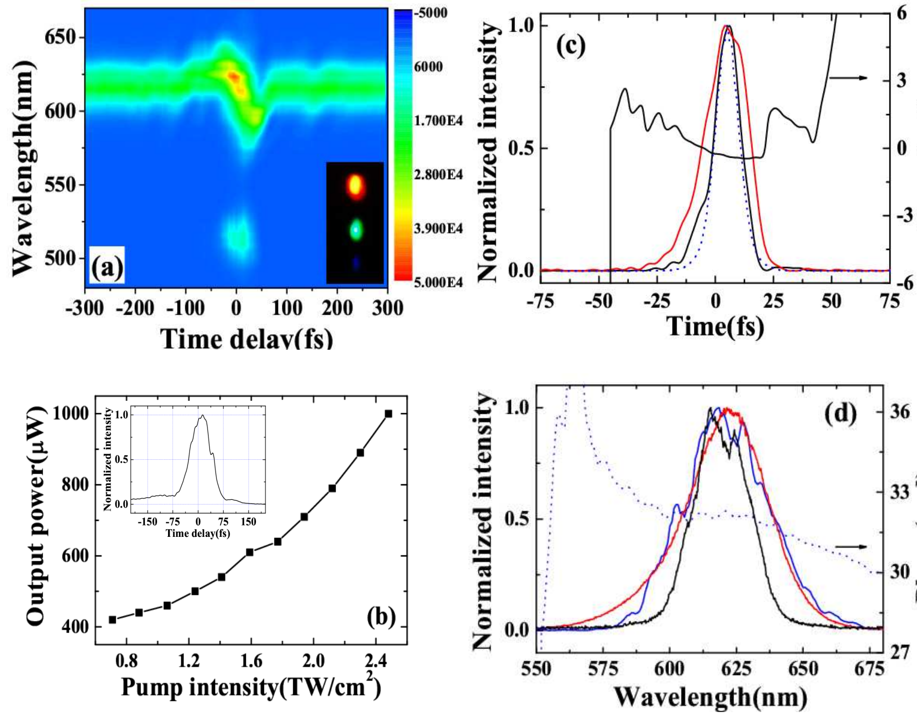

Fig. 4. (Color online) (a) The spectral profile and intensity of the output seed beam (AS3) as a function of the delay time t_{ps} when the crossing angle α was 2.80° ± 0.05°. (b) The dependence of the output energy of the output seed pulse on the pump intensity. The inset curve is the temporal profile of pump pulse. (c) The retrieved temporal profiles of the incident seed pulse (red solid line) and the compressed output seed pulse (black solid line), and the transform-limited pulse of the broadened spectrum (blue dotted line). (d) The retrieved spectrum (blue solid line) and the spectral phase (blue dotted line) of the output seed pulse. The measured spectra of the incident seed pulse (black solid line) and output seed pulse (red solid line).

This phenomenon was observed also when the first-order anti-Stokes (AS1, 620 nm) sideband [18–21] was used as the seed beam. When the crossing angle α was around 2.80° ± 0.05°, the incident seed pulse was spectrally broadened and amplified simultaneously. In this case, the incident pulse energy of AS1 was 400 nJ and the pump energy was 140 μJ. The spectral profile and intensity of the amplified output beam as a function of the delay time t_{ps} are shown in Fig. 4(a). Cascaded FWM signals were simultaneously generated in this case, as shown in the photograph in the inset of Fig. 4(a). The maximum output pulse energies of the seed beam and the first-order cascaded signal (around 500 nm) were 1.1 μJ and 250 nJ, respectively. The output energy of the seed pulse as a function of the pump intensity at a delay time of 0 fs is shown in Fig. 4(b). Much higher output energies are expected to be obtained when cylindrical lens for focusing is used [15–17]. The small thickness of the glass plate ensured that the phase-matching spectral bandwidth was broad. Therefore, there was still broadband amplification around 620 nm. In the process, the pump pulse has a slightly steeper trailing edge, which was measured by using SHG-FROG, as shown in the inset of Fig. 4(b). Furthermore, the self-steepening effect will introduce a sharper trailing edge of the pump pulse during its propagation [10]. As a result, the rapid decrease of trailing edge induced a broader blue spectral shift in the seed pulse, as shown in Figs. 3(a), 3(b) and Fig. 4(a). The steep trailing edge of the pump pulse also induced a rapid decrease of the spectral shift in the positive delay time, as is also shown in Figs. 3(a), 3(b) and Fig. 4(a).

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A quasi-linear chirp can be imposed across the weak seed pulse when the pump pulse is much wider compared with it [10]. In the same way as SPM-based compressors, the phase induced by XPM can also be compensated by using a chirped mirror pair. Furthermore, XPM based compressor can have more flexibility than SPM ones because the phase can be tuned by the pump pulse. After passing through a pair of chirped mirrors for four bounces (−40 fs^2/bounce), the pulse duration of the weak output pulse was measured by XFROG. The spectral and temporal profile and the phase were retrieved using a commercial software (FROG 3.0, Femtosoft Technologies) with a 512 × 512 grid. The retrieval error was smaller than 0.006. The retrieved temporal profiles of both the incident seed pulse and the compressed output seed pulse, and the transform-limited pulse for the broadened seed pulse are presented in Fig. 4(c). The 22.6 ± 0.5fs incident pulse was compressed to 12.6 ± 0.5 fs, which is well close to the calculated transform-limited pulse duration of 10.5 fs. The retrieved spectrum and spectral phase of the output seed pulse are shown in Fig. 4(d).

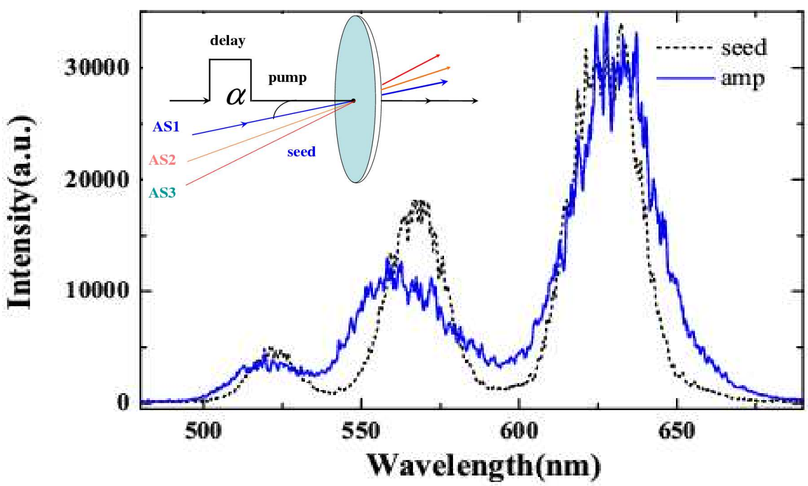

Fig. 5. (Color online) The spectral profile and intensity of AS1, AS2, and AS3 without pump (black dash line). The blue solid line is the curve of the spectral profile and intensity of AS1, AS2, and AS3 with pump. The inset was schematic of the experimental setup; α is the crossing angle between pump and AS1.

We also guided three beams, AS1, AS2, and AS3 generated by cascaded FWM [18–21] into the glass at the same time. The schematic of the experimental setup was shown in the inset of Fig. 5. The beam diameters of AS1, AS2, and AS3 on the 0.5-mm thick fused silica glass were all about 190 μm. The crossing angles between pump beam and three anti-Stokes sidebands, AS1, AS2, and AS3 were about 2.6°, 2.9°, and 3.2°, respectively. The pump pulse energy was 160 μJ with about 350μm diameter on the glass. When these seed and pump beams synchronized in time on the glass, the spectra of the three seed beams, AS1, AS2, and AS3 were broadened simultaneously, as shown in Fig. 5. The pulse energy of AS1, AS2, and AS3 were also amplified from 133nJ, 40nJ, and 12nJ to 163nJ, 50nJ, and 15nJ, respectively. This experiment proves that it is possible to simultaneously amplifying and compressing several weak pulses in a bulk medium at the same time. Then, it can be used for cascaded FWM process to optimize the spectrum of the obtained sidebands which was expected to obtain single cycle pulse after combine the optimized sidebands [22].

5. Conclusion

In summary, a novel method of simultaneously amplifying and compressing a weak pulse was demonstrated using XPM in conjunction with FWOPA in a bulk medium. It can also be used to broaden the spectra of several weak pulses with different wavelengths at the same time.

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This method is simple and can be used to optimize ultrashort pulses (for example non-collinear optical parametric amplifier system and cascaded four-wave mixing system) from UV to MIR in a broad region, which is very important for applications in photochemistry, photophysics, and photobiology. An intense tunable single-cycle pulse is expected to be obtained using this method.

Acknowledgement

We thank Dr. Kotaro Okamura for his technical assistance. This work was partly supported by the 21st Century COE program on "Coherent Optical Science" and partly supported by the grant from the Ministry of Education (MOE) in Taiwan under the ATU Program at National Chiao Tung University. A part of this work was performed under the joint research project of the Laser Engineering, Osaka University, under contract subject B1-27.

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