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# Angularly-dispersed optical parametric amplification of optical pulses with one-octave bandwidth toward monocycle regime

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**Abstract:** We demonstrated experimentally the generation of 65- $\mu$ J, 5.8-fs optical pulses with an ultrabroad bandwidth (540 – 1000 nm) by the use of a double-pass angularly-dispersed non-collinear optical parametric amplifier. We also confirmed up to the 95- $\mu$ J output from the amplifier when seed pulses were not pre-compensated for. Furthermore, we confirmed that the broadband pump pulses brought in the broader gain bandwidth (from 520 to 1080 nm) than numerical estimation based on CW-pump approximation. To the best of our knowledge, this is the system with the broadest gain bandwidth.

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**OCIS codes:** (190.4970) Parametric oscillators and amplifiers; (320.1590) Chirping.

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## 1. Introduction

Many research groups are now developing laser systems to generate extremely high-power, few-cycle pulses for high-field physics applications [1–6]. Several research groups have succeeded in the generation of isolated attosecond pulses utilizing few-cycle pulses generated by self phase modulation (SPM) in a noble-gas-filled hollow fiber [6], and utilizing multi-cycle pulses with a polarization gating technique [7]. In addition, other sophisticated techniques, such as two-color amplitude gating [8] and double optical gating [9] have been developed to generate ultrabroad XUV continuum spectra. However, in those cases, excitation sources with more than two optical cycles were employed and the durations of the generated XUV pulses were longer than 100 attoseconds. Recently, the generation of isolated sub-100-attosecond XUV pulses by the use of sub-4-fs, 300- $\mu$ J optical pulses from a hollow fiber without the above-mentioned various gating techniques has been reported [10]. This indicates that the high-power optical pulses in the monocycle regime are the most powerful candidate as a driver for the generation of shorter XUV pulses. This has been also predicted by a high-order harmonic generation theory where shorter isolated attosecond XUV pulses are generated efficiently within a half cycle of high-power, monocycle pump pulses [11, 12].

However, for the generation of shorter pulses in the monocycle regime using only chirped

mirrors, specially designed chirped mirrors with much broader bandwidth are required. But, ultrabroad-band chirped mirrors over the ultraviolet-visible-near-infrared region not only are difficult to fabricate but also have a low reflectivity ( $< \sim 90$  percent) [13] (It seems that the mirrors are not used in pulse compression experiments as far as we know). Moreover, additional dispersive optics depending on the specific optical system are required for compensation of higher-order dispersion because chirped mirrors have fixed dispersion characteristics and are unsuitable for ad-hoc precise control of the dispersion.

We have succeeded in generating 1.3-cycle, 2.6-fs optical pulses [14] by the use of SPM and induced phase modulation (IPM) in an argon-filled hollow fiber. The duration of the corresponding Fourier transform limited pulses was 2.5 fs, that is to say, the chirp of the generated pulses was almost completely compensated for by a feedback phase compensation system [15], which consisted of a M-SPIDER [16] (modified spectral phase interferometry for direct electric-field reconstruction) apparatus and a  $4-f$  pulse shaper. However, the pulse energy ( $\sim 3.6 \mu\text{J}/\text{pulse}$ ) was too low for high field physics applications due to the low throughput of the  $4-f$  pulse shaper.

Noncollinear optical parametric amplification (NOPA) has an ability to amplify broadband weak pulses and is considered to be a promising way to generate few-cycle high-power optical pulses, but its bandwidth is so narrow that only a fraction of the ultrabroad-band spectrum (460 – 1060 nm) of the pulses we have generated [15] can be amplified. For example, its gain bandwidth is limited to  $\sim 500 - \sim 800$  nm for the pump wavelength of 393 nm [17]. However, it was recently predicted by some groups [18–20] that a much broader parametric gain will be obtained if incoming seed pulses are angularly dispersed so that their frequency components almost satisfy the phase-matching condition in the gain medium in the broader wavelength region. We call this technique angularly-dispersed noncollinear OPA (A-NOPA) after this.

In this paper, we report the first experimental demonstration of ultrabroad-band optical pulse amplification with an octave bandwidth by the use of A-NOPA. We chose the second harmonic ( $\sim 400$  nm) of the output beam from a Ti:sapphire laser amplifier for pump pulses, not a Q-switched diode pumped solid state laser (DPSSL) which is generally used in an optical parametric chirped pulse amplification (OPCPA) system. The use of 400-nm pulses enables us to achieve a broader gain bandwidth. Moreover, synchronization between pump and seed pulses does not require additional low-jitter, high-speed electrical circuits.

Furthermore, we use relatively intense ultrabroad-band optical pulses generated in an argon-filled hollow fiber as a seed beam, because conventional white light continuum generated by SPM in a glass plate or a micro structure fiber is extremely weak and has a complicated spectral phase. Our hollow fiber technique does not need a high OPA gain so that we can suppress the superfluorescence in the gain medium. In addition, we have already succeeded in compensating for the chirp of the output from a hollow fiber almost completely [15, 21]. The low throughput of our  $4-f$  system which was used for the nonlinear chirp compensation [18] is not a problem because lost pulse energy will be easily recovered by an A-NOPA system. The A-NOPA system has the potential ability to generate precisely-phase-controlled, high-power optical pulses with an over-one-octave bandwidth.

## 2. Numerical calculation and design of A-NOPA system

First, we show numerically that an appropriately designed A-NOPA system has an over-one-octave bandwidth, which was already shown in Ref. [18]. We also calculated numerically the phase matching angles as a function of the wavelength for a pump wavelength of 393 nm, in the plane-wave approximation with monochromatic and undepleted pump (see Fig. 1). Here, the nonlinear crystal is a type-I  $\beta$ -barium borate (BBO). As shown in Fig. 1(b),  $\theta$  is the angle between the pump beam and the optic axis and  $\alpha$  is the noncollinear angle between the pump

and seed beams.

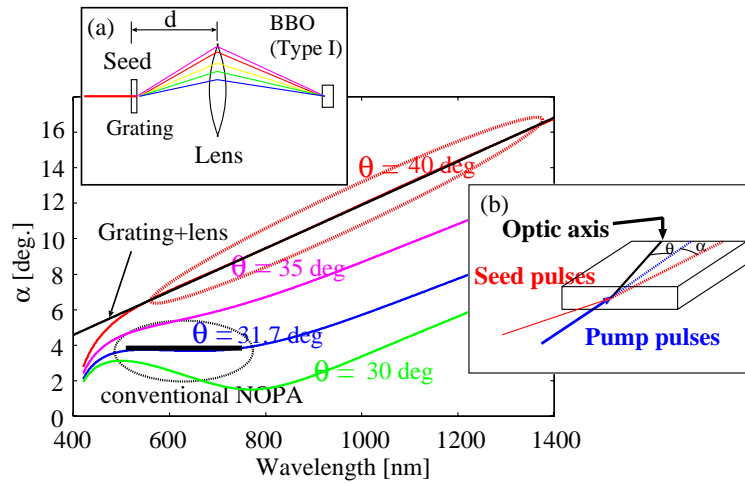


Fig. 1. Theoretical phase-matching curves for the angle  $\alpha$  between seed and pump pulses [18]. The black curve shows the angular dispersion obtained by the combination of a grating and a lens. Inset (a) basic configuration of A-NOPA and inset (b) definition of angles  $\theta$  and  $\alpha$ .

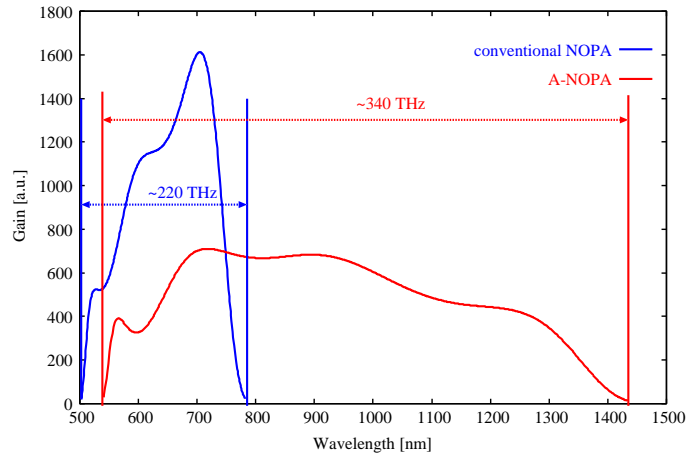


Fig. 2. Numerically calculated gain profiles of A-NOPA (red curve) and conventional NOPA (blue curve).

In conventional NOPA, the almost flat region from  $\sim 500$  to  $\sim 800$  nm is usually used when  $\theta = 31.7^\circ$ . On the other hand, when  $\theta = 40^\circ$ , there is a region where the phase-matching angle increases almost linearly to the wavelength (the red solid curve in Fig. 1) [18]. Such a linearity can be easily achieved by using a diffraction grating and a lens (or a concave mirror) as shown in Fig. 1(a). The slope of such linear dependency is easily controlled by changing the distance between the grating and the lens. For example, the black solid curve shows calculated angular dispersion when the lens (a focusing length  $f$  of 100 mm) is located at the distance  $d$  of 204

mm from the diffraction grating (a groove density of 333 lines/mm). It is found that the curve almost agrees with the phase matching angle for  $\theta = 40^\circ$  in the wavelength region from  $\sim 550$  to  $\sim 1400$  nm. Therefore, we can expect to achieve the sufficient gain in the region.

The gain profile which was numerically calculated under the same configuration is shown by a red solid curve in Fig. 2. Here, we take a pump intensity of  $50 \text{ GW/cm}^2$  and a crystal length of 1 mm. For simplicity, we assumed that the lens refocuses the seed beam dispersed by the grating but each wavelength component of the seed has no divergence. We found that the A-NOPA has an ability to amplify the ultrabroad-band pulses with an octave-spanning spectrum and the gain bandwidth is about 1.5 times broader than that of the conventional one (a blue solid curve in Fig.2). The gain of the A-NOPA is lower than that of the conventional NOPA, but it is because the gain depends on effective nonlinear coefficient  $d_{\text{eff}}$  [22], which is proportional to  $\cos \theta$  in the case of type-I BBO crystal [23]. It is not due to the configuration of the A-NOPA itself. Moreover, the pulse front of the seed beam almost matches with that of the pump in the A-NOPA configuration [18]. In our case, we numerically confirmed that the mismatch of these pulse fronts is less than several milliradian when the broadest gain bandwidth is obtained. Therefore, the pulse front tilt does not matter practically in our case.

The above simple configuration cannot be used for practical purposes. This is because we can not obtain the numerically calculated gain profile unless the divergence of each wavelength component is appropriately compensated. Figure 3 shows the practical schematic of the A-NOPA. The concave mirror CM2 (a focal length  $f_2$  of 100 mm) and the diffraction grating (a groove density of 333 lines/mm) correspond to the lens and the grating in Fig. 1, respectively. The concave mirror CM1 (a focal length  $f_1$  of 250 mm) was used to reduce the beam size to  $f_2/f_1$  and to compensate for the beam divergence by CM2. The distance between CM1 and CM2 was set to  $f_1 + f_2$ . The beam angularly-dispersed by these components, which goes through the gain medium (1-mm-thick type-I BBO crystal), is recombined by the symmetrically placed optical components (see Fig. 3) to get a single collimated beam [24]. We confirmed ultrabroad-band gain from 550–1000 nm (output pulse energy of  $7.8 \mu\text{J}$ ) by preliminary experiments using this configuration.

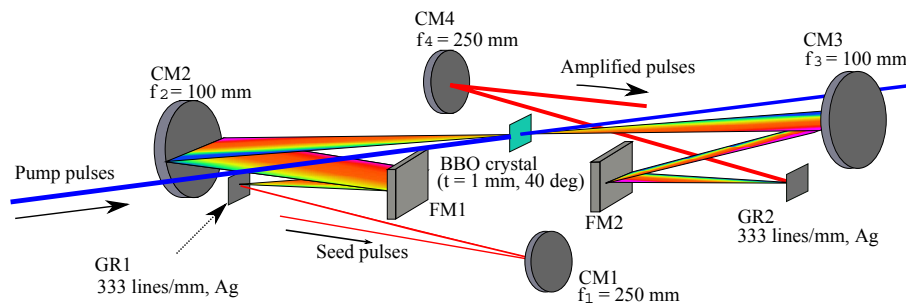


Fig. 3. Schematic of angularly-dispersed NOPA (single pass geometry). CM1, CM2, CM3, CM4: silver-coated concave mirrors, FM1, FM2: silver-coated folding mirrors, GR1, GR2: silver-coated diffraction gratings

Furthermore, to get higher pulse energy, we designed a new A-NOPA which enables twice the amplification. As shown in Fig. 4, the angularly-dispersed pulses were symmetrically reflected back by the use of the concave mirror CM3 ( a focal length  $f'_3$  of 250 mm) and folding mirrors when the optical path length between BBO crystal and CM3 and that between CM3 and FM3 were set to equal to  $f'_3$ . Therefore, the pulses were amplified again by the reflected pump pulses. In both configurations, the seed pulses were reflected twice by the diffraction gratings. The

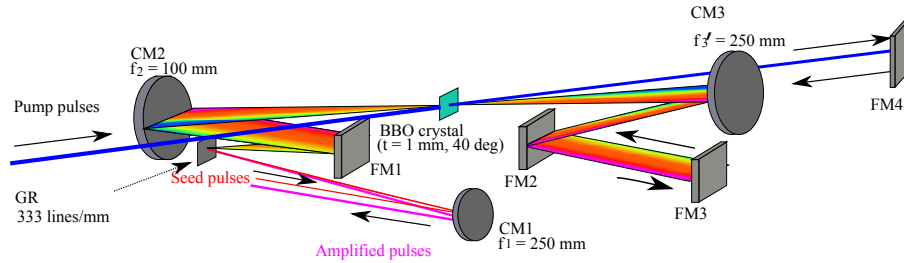


Fig. 4. Schematic of angularly-dispersed NOPA (double pass geometry). CM1,CM2,CM3: silver-coated concave mirrors, FM1,FM2,FM3: silver-coated folding mirrors, FM4: dielectric mirror, GR: silver-coated diffraction grating

loss in the A-NOPA system was mainly caused by diffraction gratings. Therefore, the new configuration shown in Fig. 4 is superior to the previous one in the point that it enables twice the amplification without increasing the loss.

### 3. Experiment and discussion

Our experimental setup is shown in Fig. 5. We let the split beam ( $300 \mu\text{J}/\text{pulse}$ ) of the output from a cryogenically-cooled Ti:sapphire laser amplifier system (Red dragon II, KMLabs inc., a pulse duration of 25 fs, a center wavelength of 786 nm, a repetition rate of 1 kHz) propagate in a hollow fiber (a length  $l$  of 370 mm, an inner diameter  $d$  of 0.1 mm) which was set in a chamber filled with 3.0-atm argon gas.

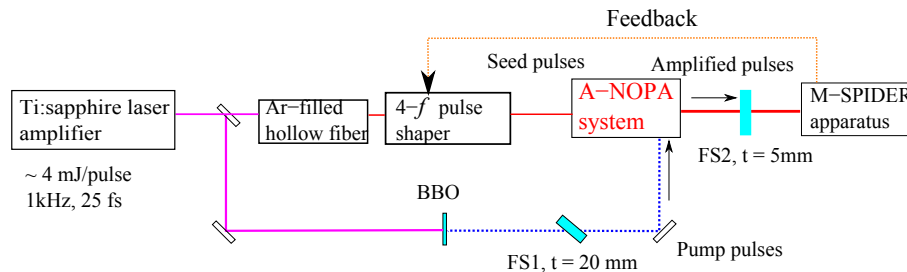


Fig. 5. Schematic of experimental set-up. FS1, FS2: Fused silica plate, BBO: Type-I BBO crystal ( $t = 0.2 \text{ mm}$ ,  $\theta = 40 \text{ deg}$ ).

Ultrabroad-band pulses generated by SPM in the noble gas were guided into a grating-based  $4-f$  pulse shaper with a liquid crystal spatial light modulator (SLM, a pixel number of 648, a transmission range of 400 - 1300 nm, a pixel size of  $102 \mu\text{m}$ ) [14, 15, 21], and subsequently into the A-NOPA system as down-chirped seed pulses (a pulse energy of  $\sim 0.5 \mu\text{J}$ , a spectrum from 500 to 1100 nm, a pulse duration of  $\sim 300 \text{ fs}$ ), as shown in Fig. 4. The input pulses were spatially and temporally overlapped with pump pulses (a center wavelength of 393 nm, a pulse energy of 1.2 mJ/pulse, a pulse duration of  $\sim 400 \text{ fs}$ ) which were generated by frequency-doubling of the output (pulse energy of 2.8 mJ/pulse) from the Ti:sapphire laser amplifier using a 0.2-mm-thick type-I BBO crystal. The 393-nm pump pulses were temporally stretched by a 20-mm-thick fused silica plate (FS1) to avoid spontaneous optical parametric generation in the amplifying medium (type-I BBO, a thickness of 1 mm,  $\theta = 40 \text{ degrees}$ ). The diameter of the pump beam on the BBO crystal was  $\sim 2 \text{ mm}$ , which was almost same as the seed. The

intensity of the up-chirped pump beam was  $\sim 75 \text{ GW/cm}^2$ . The seed pulses were negatively chirped by the  $4-f$  pulse shaper before optical parametric chirped pulse amplification, and the chirp of the amplified pulses was compensated for by a 5-mm thick fused silica plate. Then, the chirp-compensated pulses were guided into the M-SPIDER apparatus for the measurement of the spectral phase. The residual chirp was further compensated for by the  $4-f$  pulse shaper.

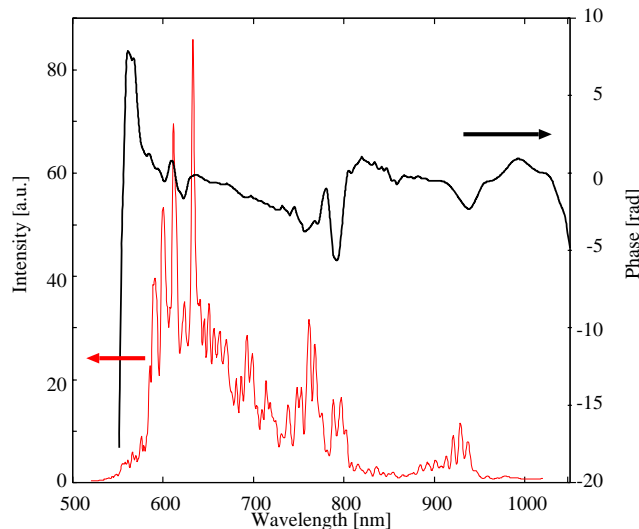


Fig. 6. Spectrum of amplified pulses and corresponding reconstructed spectral phase

We confirmed an ultrabroad-band amplification with nearly one-octave bandwidth ranging from  $\sim 550$  to  $\sim 1000$  nm, as shown in Fig. 6. The sensitivity of CCD was calibrated by using a calibrated light source. We found that the spectral phase is almost flattened, but higher order dispersion still remains. This is because the modulation of the spectrum made reconstruction of the spectral phase by SPIDER difficult and therefore, we could not compensate for the residual chirp with high accuracy. The output pulse energy, which was reduced by about 50 percent mainly due to diffraction losses by the gratings, was  $65 \mu\text{J/pulse}$ . Moreover, it should be noted that we also could boost pulse energy up to  $95 \mu\text{J/pulse}$  when the LC-SLM was not activated.

Figure 7 shows the reconstructed temporal profile of amplified pulses. We found that the temporal profile of amplified pulses has little sub pulses and the duration of the main pulse is 5.8 fs, while the duration of the corresponding Fourier transform limited pulses is 4.0 fs.

Finally, we investigated the possibility of the further enlargement of the gain bandwidth by a broadband pumping beam. Figure 8 shows the calculated curves of gain profiles for different wavelengths of pump beams using the same configuration. We found that gain profile drastically changes with a slight shift in the pumping wavelength: the shorter wavelength shift extends both edges of the gain profile. We produced a 10-nm-FWHM (Full Width of Half Maximum) bandwidth pumping beam using a thin BBO crystal (a thickness of  $200 \mu\text{m}$ ). If both the seed and pump pulses are appropriately chirped, it can be expected that an enlarged gain profile will be obtained in  $\sim 520 - \sim 1500$  nm. In fact, we confirmed ultrabroad-band amplification in 520 – 1080 nm by using the 10-nm bandwidth pump pulses and applying the phase modulation to the seed pulses by the pulse shaper in order to obtain the broader gain bandwidth (see Fig. 9). To the best of our knowledge, this is the NOPA system which has the broadest gain bandwidth. However, further optimization is required to achieve a good balance between broadband

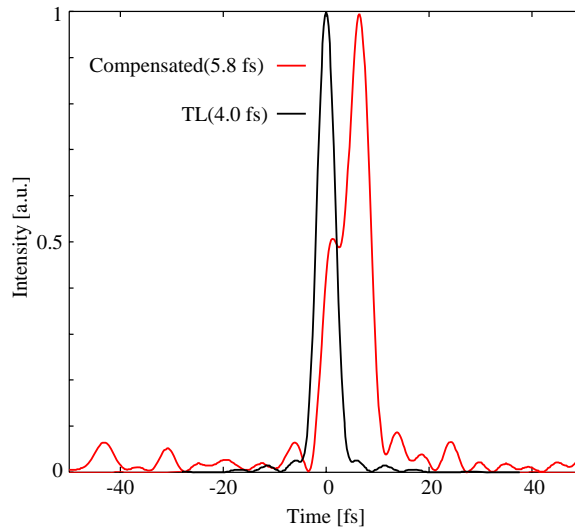


Fig. 7. Temporal profiles of reconstructed and the corresponding Fourier-transform-limited pulses.

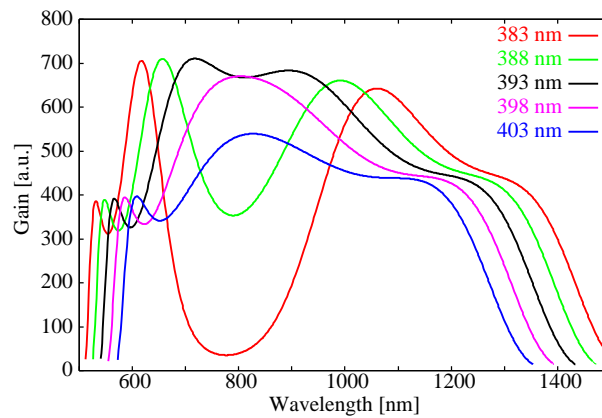


Fig. 8. Curves of calculated gain profiles. All the curves are calculated under the same conditions without wavelength of pumping beam.

amplification and pulse compression.

#### 4. Conclusion

It was demonstrated experimentally that A-NOPA is an attractive way to obtain octave-spanning broadband (520–1080 nm) pulses with high pulse energy in the monocycle region. We also could boost the seed pulses up to 95  $\mu\text{J}/\text{pulse}$ , and confirmed the generation of 5.8-fs, 65  $\mu\text{J}/\text{pulse}$  by using a pulse shaper.

However, in order to obtain higher-power shorter optical pulses, there are some problems to be solved in our present system. The first is the intensity modulation in the amplified spectrum, and the second is the optical damage on the diffraction grating. These problems are not caused

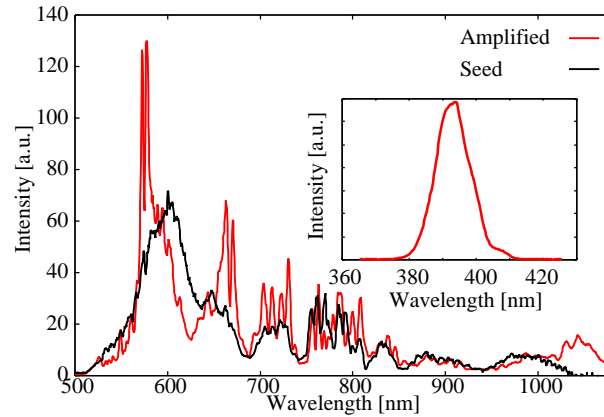


Fig. 9. The spectra of amplified pulses and the corresponding seed pulses. Inset shows the spectrum of pump pulses.

by the intrinsic limitation of the A-NOPA but is due to the design of the optical system. We are considering the following solutions.

As for the former, the modulation is due to the SLM, in addition to intrinsic characteristics of SPM. The negative GDD added by SLM for chirped pulse amplification was about  $-550 \text{ fs}^2$ . This value slightly exceeds typical limitation ( $-400 \text{ fs}^2$ ) for our  $4-f$  system, which depends on “load per pixel” [21]. Too much load not only decreases the accuracy of pulse compression, but also causes the intensity modulation by diffraction on SLM. This problem can be solved by the use of additional dispersive optics such as a prism pair and chirped mirrors.

The latter arises from the fact that the beam size (diameter of  $\sim 2 \text{ mm}$ ) on the diffraction grating is approximately the same as that on BBO crystal in the configuration shown in Fig. 4. Actually, the grating we used could not bear amplified pulses with more than  $95 \mu\text{J}/\text{pulse}$ . We are now developing an improved A-NOPA whose damage threshold is enhanced more than 5 times. Several-hundred micro-joule-level pulses in the monocycle regime will be generated in the near future.

In principle, the A-NOPA has the disadvantages that the gain is lower than the conventional one and the output pulse energy is partially lost due to the diffraction loss by a grating. However, at present, only A-NOPA can amplify the optical pulses with octave-spanning bandwidth in the monocycle regime. The A-NOPA will become the very promising candidate as a driver for application such as the generation of shorter attosecond pulses.