# Sub-15fs ultraviolet pulses generated by achromatic phase-matching sum-frequency mixing

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**Abstract:** A broadband ultraviolet pulse with a spectral width of 44 nm was generated by achromatic sum-frequency mixing of an 805-nm pulse and ultrabroadband visible pulse. Angular dispersion was introduced to achieve broadband phase matching by a prism pair. The UV pulse was compressed to 13.2 fs with another prism pair, with energy of 600 nJ.



**OCIS codes:** (140.4480) Optical Amplifiers; (190.4410) Nonlinear Optics and Parametric Processes; (190.4970) Parametric Oscillators and Amplifiers

## References and links

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## 1. Introduction

Ultrashort broadband ultraviolet (UV) laser pulses are an important tool for investigating elementary processes in physics, chemistry, and photobiology [1]. Considerable effort has been expended to generate ultrashort broadband UV pulses. The main difficulty in achieving ultrashort UV pulses is obtaining broadband UV spectra. Short UV pulses have been generated in air or in rare gasses either by the third harmonic of a Ti: sapphire laser [2], or by

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parametric difference-frequency mixing between the fundamental frequency and its second harmonic, which has been performed in a hollow fiber [3,4] and in a filament [5]. A sub-20-fs UV pulse was directly generated by second harmonic generation (SHG) of a broadband visible pulse [6], using a 50-um-thick BBO crystal to increase the phase-matching bandwidth. Ultrashort broadband UV pulses were generated by SHG, using achromatic phase matching to circumvent the bandwidth limitations [7,8]. Nabekawa et al.[9,10] proposed and demonstrated a sum-frequency mixing, using achromatic phase matching to generate a broadband UV with a spectral width of ~10 nm in the wavelength region of ~256 nm, with a broadband infrared pulse and the SHG of the infrared pulse with a narrow spectral width. The generated UV is compressed to sub-20-fs, using a pair of prism and a liquid-crystal spatial light modulator (LC-SLM). This system looks a little complex.

In this paper, we generated broadband UV pulses by achromatic phase-matching sum-frequency mixing (ASFM) of a femtosecond pulse with a central wavelength of 805 nm and an ultrabroadband visible pulse from a two-stage noncollinear optical parametric amplification (NOPA) system. A prism pair was employed to introduce the required angular dispersion, and another prism pair was used to compress the broadband UV pulses and reduce the spatial chirp. The compressed UV pulses had a pulse width of 13.2 fs with energy of 600 nJ.

## 2. Experiment

Figure 1 shows a schematic diagram of the NOPA and UV setup. A Bright chirped pulse amplifier system was used as the source, delivering 130-fs pulses with energy of 1.4 mJ at 805 nm and a repetition rate of 1 kHz. For generating broadband NOPA, a small fraction of the output energy from the CPA laser was used to generate a white light as seed in a 2-mm-thick sapphire crystal. The seed was then precompressed by two parallel gratings with 150 *l*/mm, separated by a distance of 12 mm. The long wavelengths of the white light were cut off by a 1-mm-thick high-pass filter. Figure 2 shows the spectrum of the seed. Then the seed was amplified by the two stages of NOPA system, using two 1-mm-thick BBO crystals with cutting angle of 31.5$^{o}$. The amplified pulse energy of the first NOPA stage is 0.5 μJ, with pump energy of 10 uJ. The output pulse energy of the second stage is 5.0 μJ, with pump energy of 25 uJ, and the spectrum of the visible pulse is also shown in Fig. 2.

The configuration for ASFM of the visible pulse and the fundamental pulse to generate broadband UV is shown in Fig. 3. The visible pulses from the two NOPA stages are dispersed by two fused silica prisms with apex angles of 68.9$^{o}$. The first prism disperses the visible pulse and the second prism converts the angular dispersion into a lateral dispersion. The incident angles of NOPA on the surfaces of the prisms are at the Brewster angle, and the distance *d* of the two prisms is about 670 mm to maximize the phase-matching bandwidth. Because the polarization of visible pulse is vertical, therefore the energy loss is high when it passes through the prisms. Thus, the energy of the visible pulse before entering the crystal of ASFM is 2.4 uJ. The resulting collimated visible beam having spatially displaced frequency components is focused onto a common spot by a 90$^{o}$ parabolic mirror with *f*=50 mm for ASFM. The fundamental pulse for ASFM is stretched by 100-mm-thick bulk silica to about 240 fs, in order to make the fundamental pulse width matching the pulse width of NOPA and increase the energy of generated UV, with energy of 70 uJ. The 805 nm fundamental pulse was focused by a 400-mm focal lens. A 1-mm-thick BBO crystal is used for ASFM, with type I phase matching and cutting angle of 43$^{o}$. The angle outside the BBO crystal between the ~650 nm portion of the visible pulse beam and fundamental pulse beam is about 20$^{o}$, which corresponds to an inside angle of about 12$^{o}$. And the propagation direction of the fundamental pulse beam was normal to the surface of the BBO crystal, which was located before the focal point of the NOPA. The generated UV pulse was collimated by a concave mirror with f=100 mm. It was then compressed by a pair of fused silica prisms with apex angles of 68.9$^{o}$. The dispersion of UV could be adjusted by varying the insert depth of the first compression prism. The energy loss was minimal because the polarization of UV was horizontal and the incident angle of the UV pulse on the prism surface was almost at the Brewster angle.

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The broadest spectrum of UV pulse is shown in Fig. 5. We also simulated the phase mismatching of the ASFM by optical ray tracing, using the same parameters as those given above [10]. The residual phase mismatching of the ASFM setup at three different prism distances *d* (665 mm, 670 mm and 675 mm) is shown in Fig. 4. In Fig. 4, the bottom and top horizontal axes in the figure represent the wavelengths of the UV spectrum and the corresponding NOPA spectrum, respectively. And it shows the phase mismatching is not very sensitive to the prism distance *d*. The generated broadest spectra of UV are generally consistent with the simulation result, compared with NOPA and UV spectra.

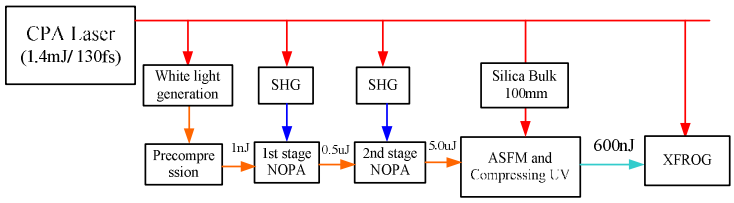



Fig. 1. Schematic diagram of the NOPA and UV setup

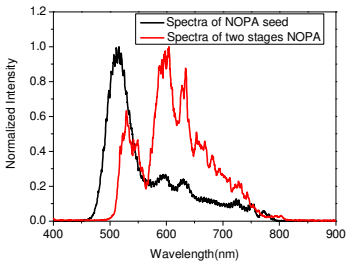
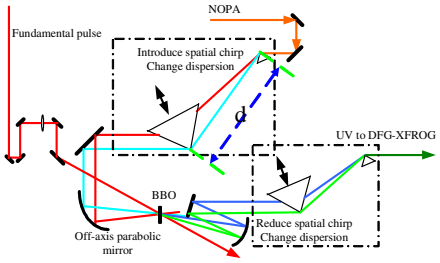



Fig. 2.The seed and NOPA output spectra




Achromatic phase-matching
sum frequency mixing (ASFM)

Fig. 3. Configuration of ASFM and compression of UV

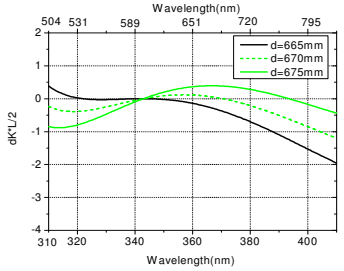



Fig. 4. Residual phase mismatching of the ASFM setup at three different prism distances, with a 1-mm-thick BBO crystal.

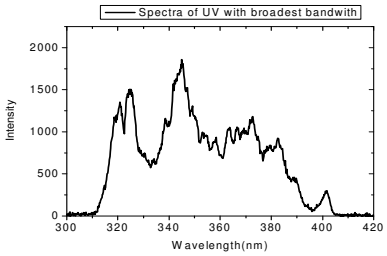



Fig. 5. The broadest bandwidth UV spectrum obtain by ASFM, with d=670mm and L=1mm BBO crystal.

Then we maximized the UV pulse energy by reducing the noncollinear angle between the visible beam and the fundamental beam and finely adjusting the crystal angle of the BBO. By doing this we achieved pulse energy of 600nJ after compression. The corresponding spectra are shown in Fig. 6. And the spatial chirp of UV is simultaneously reduced by the compression prism pair. After compression, the UV beam propagated with no noticeable angular dispersion. The pulse width of the UV pulse was optimized by varying the insert depth of the prism. The temporal pulse shape of the UV pulses was measured using a homemade difference frequency generation cross-correlation frequency resolved optical gating (XFROG) [11]. The reference pulse is the fundamental pulse with pulse width of 130 fs. A 30-um-thick BBO crystal is used for XFROG, with type I phase matching and a cutting

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angle of 40°. The retrieved UV temporal shape and temporal phase are shown in Fig. 7, with the temporal width of 13.2fs (FWHM). The retrieved UV pulse shape reveals that there exist higher-order dispersions.

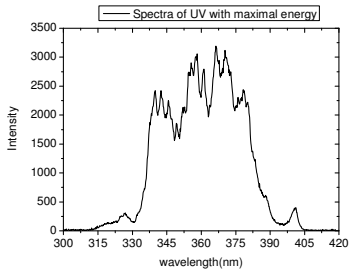


Fig. 6. Measured spectrum of UV with maximal energy.

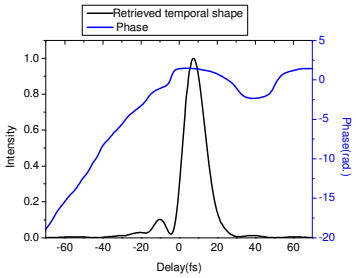


Fig. 7.The retrieved temporal shape and phase of UV with XFROG.

## 3. Conclusions

13.2-fs broadband UV pulse with energy of 600 nJ has been obtained by ASFM using a BBO crystal. A prism pair and an off-axis parabolic mirror were used to achieve achromatic phase-matching to obtain broadband UV. Such ultrashort UV pulses are ideal sources for broadband UV detection in pump-probe experiments.


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