Widely tunable sub-30 fs ultraviolet pulses by chirped sum frequency mixing

Ida Z. Kozma, Peter Baum, Stefan Lochbrunner and Eberhard Riedle

Lehrstuhl für BioMolekulare Optik, Sektion Physik, Ludwig-Maximilians-Universität München, Oettingenstrasse 67, 80538 München, Germany
Ida.Kozma@physik.uni-muechen.de

http://www.bmo.physik.uni-muenchen.de

Abstract: A novel scheme for the generation of UV pulses in the 295 - 450 nm range is presented. Sum frequency mixing of the chirped visible pulses from a noncollinear optical parametric amplifier with deliberately chirped pulses from the Ti:sapphire amplifier ensures efficient energy conversion and easy tunability. Pulse energies as high as 5.5 μJ at 295 nm, and >2 μJ in most of the tuning range are obtained with highly symmetric and smooth spectra. They are compressed to sub-30 fs throughout the entire tuning range (20 fs at 348 nm) with a newly designed prism compressor.

©2003 Optical Society of America

OCIS codes: (320.0320) Ultrafast optics; (190.4970) Parametric oscillators and amplifiers; (190.2620) Frequency conversion; (320.7080) Ultrafast devices

---

References and links

1. T. Wilhelm, J. Piel, E. Riedle, "Sub-20-fs pulses across the visible from a blue-pumped single-pass noncollinear parametric converter," Opt. Lett. **22**, 1494-1496 (1997).
2. E. Riedle, M. Beutter, S. Lochbrunner, J. Piel, S. Schenkl, S. Spörlein, W. Zinth, "Generation of 10 to 50 fs pulses tunable through all of the visible and the NIR," Appl. Phys. B **71**, 457-465 (2000).
3. P. Baum, S. Lochbrunner, L. Gallmann, G. Steinmeyer, U. Keller, E. Riedle, „Real-time characterization and optimal phase control of tunable visible pulses with a flexible compressor," Appl. Phys. B **74** [Suppl.], S219-S224 (2002).
4. G. Cerullo, S. De Silvestri, "Ultrafast optical parametric amplifiers," Rev. Sci. Instrum. **74**, 1-17 (2003).
5. K.R. Wilson, V.V. Yakovlev, "Ultrafast rainbow: tunable ultrashort pulses from a solid-state kilohertz system," J. Opt. Soc. Am. B **14**, 444-448 (1997).
6. L.D. Ziegler, J. Morais, Y. Zhou, S. Constantine, M.K. Reed, M.K. Steinershepard, D. Lommel, "Tunable 50-fs pulse generation in the 250-310-nm ultraviolet range," IEEE J. Quantum Electron. **34**, 1758-1764 (1998)
7. A.J. Wurzer, S. Lochbrunner, E. Riedle, "Highly localized vibronic wavepackets in large reactive molecules," Appl. Phys. B **71**, 405-409 (2000).
8. A. Kummrow, M. Wittmann, F. Tschirschwitz, G. Korn, E.T.J. Nibbering, "Femtosecond ultraviolet pulses generated using noncollinear optical parametric amplification and sum frequency mixing," Appl. Phys. B **71**, 885-887 (2000).
9. A. Tzankov, T. Fiebig, I. Buchvarov, "Tunable femtosecond pulses in the near-ultraviolet from ultrabroadband amplification," Appl. Phys. Lett. **82**, 517-519 (2003).
10. Y. Nabekawa, K. Midorikaawa, "Broadband sum frequency mixing using noncollinear angularly dispersed geometry for indirect phase control of sub-20-femtosecond UV pulses," Opt. Express **11**, 324-338 (2003), http://www.opticsexpress.org/abstract.cfm?URI=OPEX-11-4-324.
11. A.C.L. Boscheron, C.J. Sauteret, A. Migus, "Efficient broadband sum frequency based on controlled phase-modulated input fields: theory for 315-nm ultrabroadband or ultrashort pulse generation," J. Opt. Soc. Am. B **13**, 818-826 (1996).

---

1. Introduction

Ultrashort pulses applied in pump-probe experiments allow one to study physical and chemical processes with highest temporal resolution. Parametrically generated pulses have

broad frequency tunability in the near infrared (NIR) and visible spectral region and the noncollinear geometry provides ultrabroadband pulses [1-3]. As a result, noncollinearly phase-matched optical parametric amplifiers (NOPAs) became a widespread source of tunable ultrashort pulses [4]. Most organic molecules have their electronic absorption in the blue and ultraviolet (UV) spectral region. Therefore it is essential to extend the available spectral range to shorter wavelengths and to develop schemes for efficient generation of ultrashort optical pulses tunable in the blue and UV.

Tunable UV pulses were generated from parametrically generated pulses by frequency doubling, quadrupling or upconversion with a powerful NIR pulse [5]. The tunability of pulses obtained from second harmonic generation (SHG) ranges from 250 nm to 350 nm, with pulse energies up to 120 nJ [6]. The shortest pulse duration reported so far was 24 fs [7]. Essentially transform limited 33-fs pulses in the range from 315 nm to 355 nm with an energy level of 1-2 µJ were achieved using sum frequency mixing (SFM) of visible pulses from a NOPA with pulses of a sub-40-fs Ti:sapphire amplifier [8]. Alternatively, broadband phase matching in a 258-nm-pumped NOPA directly delivers pulses with a duration of 24-35 fs in the range of 335-480 nm with energies of a few hundred nJ [9]. A SFM scheme using a noncollinear and angularly dispersed geometry was reported to produce pulses with a broad spectrum around 320 nm and 1.3 µJ pulse energy [10]. Pulses generated with this method are not inherently tunable and have not yet been compressed.

In this paper, we present a scheme for SFM that provides pulses of some µJ energy with an unprecedented tuning range from 295 nm to 450 nm. By mixing chirped pulses we achieve pulse energies as high as 5.5 µJ at 295 nm, and more than 2 µJ in most of the tuning range. Their spectral profiles are highly symmetric and smooth. We designed a UV prism sequence and compressed the pulses to 20 fs at 348 nm, and below 30 fs throughout the entire tuning range.

2. Chirped SFM

Figure 1 compares the wavelength ranges available with the SHG and SFM schemes for the given Ti:sapphire chirped pulse amplifier (CPA) wavelength and NOPA tuning range. Around the wavelength of the CPA, from 700 nm to 870 nm there is a gap due to the highly structured spectrum of the seed continuum in this range. A first advantage of the SFM scheme is that in the UV the resulting gap around 400 nm is only half of that found in SHG. The second benefit is that larger pulse energies can be achieved from the SFM process. A third advantage is that fluctuations of energy and beam profile of the parametrically generated pulse are not increased, since SFM only adds the much smaller fluctuations of the CPA to the overall response [8]; therefore, a higher stability is achieved.
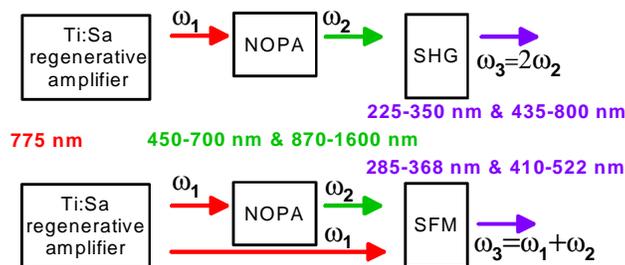

Fig. 1. Second harmonic generation and sum frequency mixing schemes based on parametric devices for the generation of tunable UV pulses.

As source of continuously tunable pulses in the visible and the NIR, we use a two-stage NOPA (shown schematically in Fig. 2(a)), in which the output of the first parametric amplifier is used as the seed light of a second amplification stage. The spectral width, that is the transform limited pulse length of the NOPA output, can be adapted to the experimental needs by varying the chirp of the seed continuum [2]. So far only the compressed NOPA pulse

lengths were reported [2]. In order to set up an efficient mixing scheme, it is of fundamental importance to know the temporal characteristics of the uncompressed NOPA output.
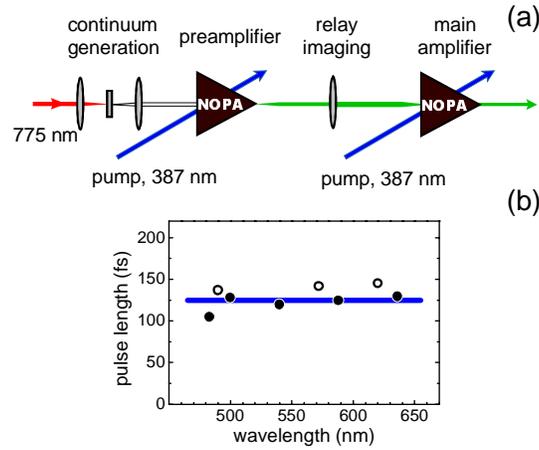

Fig. 2. (a) Principal parts of the two-stage NOPA. (b) Pulse duration of the uncompressed NOPA output. A lens having higher (●) or lower material dispersion (○) is used for collimating the continuum. The blue line denotes the duration of the frequency doubled pump pulses.

For cross correlation measurements, an additional one-stage NOPA is tuned to 600 nm and the pulses are compressed in a fused silica prism compressor to 25 fs. The correlation between the outputs of the two NOPAs is measured in a 25-μm-thick BBO crystal while the two-stage NOPA is tuned from 480 nm to 640 nm. The deconvoluted pulse lengths of the uncompressed NOPA pulses are shown in Fig. 2(b). The pulse duration shows only a small variation around 125 fs in the full visible wavelength range and is independent of the dispersion introduced to the seed continuum by inserting dispersive material. This value corresponds to the duration of the frequency doubled pump pulses. The duration of the NOPA output pulse is primarily determined by the length of the pump pulse independent of the central wavelength and the spectral width of the output.
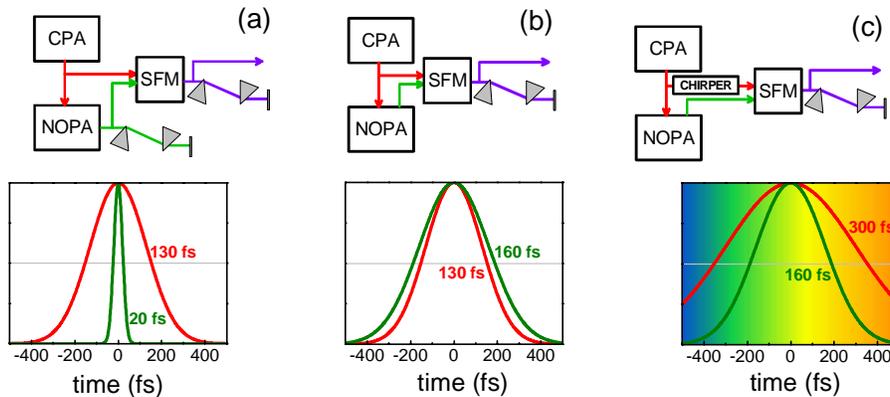

Fig. 3. Three schemes for SFM. (a) Compressed NOPA pulses requiring two compressors, (b) uncompressed NOPA pulses with one compressor, and (c) chirped SFM of stretched NIR pulses and uncompressed NOPA pulses. The graphs below illustrate the temporal overlap between the NIR and the NOPA pulses.

Possible schemes for SFM arrangements of NIR pulses from the CPA with visible NOPA pulses are compared in Fig. 3. We assume 130-fs-long NIR pulses and that the visible pulses from the NOPA, which can be compressed to a duration of 20 fs, are lengthened to 160 fs during propagation through the optical system.

Naïvely, one assumes that the SFM of well compressed pulses should render the shortest possible UV pulses. The arrangement (shown in Fig. 3(a)) has an obvious shortcoming. In real spectroscopic applications, a second compressor is needed after the SFM crystal to compensate for the group velocity dispersion of the various optical elements in the UV beam path. The simultaneous adjustment of two prism compressors makes the tuning tiresome when the central wavelength of the NOPA is changed. Furthermore, the temporal overlap between the interacting pulses is poor, which results in an inefficient energy transfer.

A more flexible arrangement is the one shown in Fig. 3(b); here the uncompressed NOPA pulse interacts with the fundamental pulse, so only one prism compressor is needed for dispersion compensation. Unfortunately, however, the duration of the visible pulse is slightly longer than that of the NIR pulse, so parts of the visible pulse are not converted in the SFM process. The NOPA pulse is strongly chirped, therefore, the extreme spectral components are lost, resulting in a narrower bandwidth. The seemingly improved temporal overlap results in pulses having less energy and longer duration.

To achieve efficient broadband frequency conversion, the NIR pulse can be chirped in a glass block and stretched to a duration which results in a good temporal overlap (see Fig. 3(c)). The arrangement takes advantage of the full broad visible spectrum and ensures efficient energy transfer. At the same time, wavelength tuning remains simple, since only one compressor is necessary and the same stretcher is adequate for the entire tuning range. A theoretical treatment of chirped frequency mixing is given in ref. [11]. For SFM scheme discussed in this work the direction of the chirp does not play a major role since the NOPA bandwidth is many times that of the CPA.

3. Experimental setup

Our experimental setup is shown in Fig. 4. A Clark MXR CPA 2061 system is used as pump laser delivering 130-fs pulses with 1 mJ energy at 775 nm and 3 kHz repetition rate. Of this energy, ~250 μJ is used to pump the two-stage NOPA. The resulting tunable 2 to 14-μJ pulses are collimated and focused ($f = 200$ mm) by reflective optics. A fraction of ~50 μJ of the 775-nm pulse is split off and sent through a glass block. We verified, through calculations, that a 45-mm-long SF57 slab is suitable to create the necessary chirp of the fundamental pulse. This stretched pulse (about 280 fs calculated duration) is focused with an $f = 300$ mm
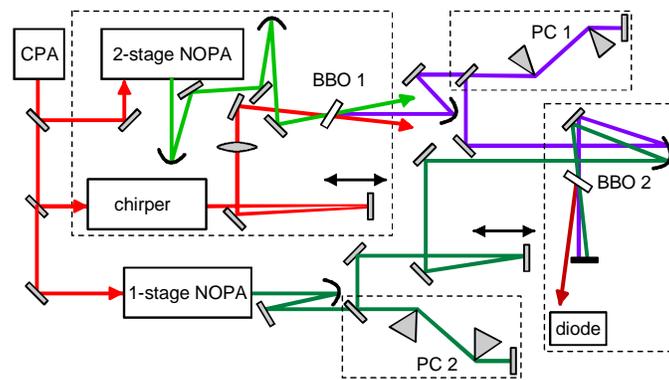

Fig. 4. Experimental setup for SFM. The uncompressed output of the 2-stage NOPA is upconverted with the stretched output of the CPA (BBO 1). The generated UV pulse is compressed in a UV prism compressor (PC1). A 1-stage NOPA delivers visible pulses, which are compressed to about 20 fs duration (PC2). The cross correlation of the UV and the visible pulses (BBO 2) is detected with a Si photodiode.

lens and overlapped with the NOPA pulse in a type-I BBO crystal. The focal planes of both beams are ~10 mm before the crystal surface to avoid deterioration of the generated beam profile due to nonlinear propagation effects and possible crystal damage. Crystals with cut angles of 23°, 30°, 35° and 40° are used to ensure efficient phase matching at different wavelengths. The nominal thickness of the crystals are 100 μm, which is a compromise between attainable energy and the duration of the generated UV pulse. Broader spectra should be obtainable using thinner, higher pulse energies using thicker crystals for SFM. The pulses are mixed in a slightly noncollinear geometry to spatially separate the SFM signal from the weak ones generated by doubling the individual pulses; the beam intersection angle is about 1.5°. To collimate the SFM signal an aluminum mirror with a focal length of 200 mm is used.

For compression of the UV pulses to a minimum duration a double-pass prism pair is used. Usually in prism compressors the apex angle is cut such that the angle of incidence is the Brewster angle at the central wavelength. Thus, the Fresnel reflection losses for *p*-polarized light are minimized and the system is essentially loss free. Since the SFM signal is vertically polarized, a special UV prism compressor was designed to keep the plane of the compressor parallel to the optical table without the need for rotating the polarization of the UV light. We chose fused silica prisms with an apex angle of 45°. This results in an angle of incidence of about 35° for the UV pulses. For this moderate angle of incidence a broadband anti-reflection coating for *s*-polarized light is available (BBAR240-420/35s; LASER COMPONENTS GmbH), which reduces the loss per surface to 2% in the 230-460 nm region and to less than 1% in the 240-430 nm region. The typical spacing of the prisms varies between 50 cm and 100 cm. A dielectric mirror is used as end mirror.

For the temporal characterization of the compressed UV pulses difference frequency cross correlations are measured in a 100-μm-thick BBO crystal with the output of an additional one-stage NOPA, which is compressed in a prism sequence with Brewster's angle fused silica prisms.
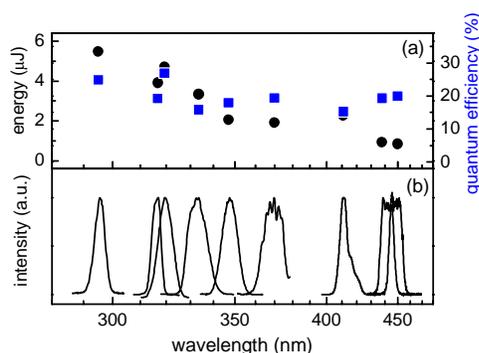

Fig. 5. Characteristics of the generated UV pulses: (a) output energy (●) and quantum efficiency (■) of the SFM process defined as the ratio of the number of generated UV photons and the visible photons and (b) spectra of the pulses.

4. Results

The characteristics of the UV pulses generated by chirped SFM are presented in Fig. 5. The tuning range is spanning from 295 nm to 450 nm. The spectra are smooth and symmetric as shown in Fig. 5(b). The quantum efficiency of the SFM process is about 20 %. The maximum output energy of 5.5 μJ was obtained at the shortest wavelength, and the pulse energy is decreasing with increasing wavelength. This tendency corresponds well to the output energy values of the NOPA. The pulse energies are measured after the SFM crystal. In supplementary measurements we verified that the total loss of UV energy while propagating through the prism compressor is 12 % or less.

Our system is optimized not only for maximum output power, but also for shortest pulse duration. At the cost of beam profile quality and spectral width higher output energies might be possible with a thicker SFM crystal or tighter focusing. The duration of the UV pulses was measured at four characteristic points of the tuning range. The observed pulse lengths are summarized along with the corresponding spectra in Fig. 6. The deconvoluted pulse lengths are obtained assuming $sech^2$ pulse shape, and vary from 19.9 fs to 28.0 fs. The time-bandwidth product varies from 0.40 to 0.54.
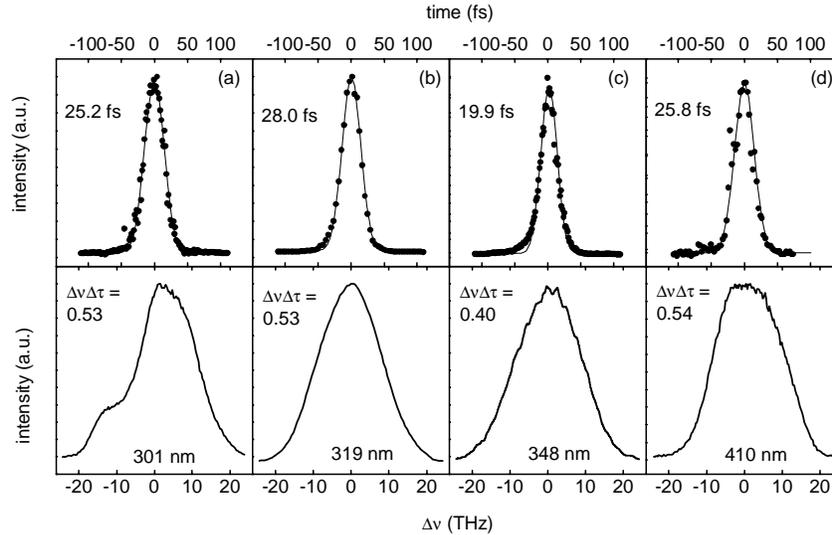

Fig. 6. Cross correlation traces and measured spectra. (a) For pulses at $\lambda = 301$ nm the spectral width is $\Delta\lambda = 6.3$ nm. With visible probe pulses of $\Delta\tau = 21$ fs the width of the cross correlation trace is $\Delta t = 35.6$ fs; (b) $\lambda = 319$ nm, $\Delta\lambda = 6.5$ nm, $\Delta\tau = 14.7$ fs, $\Delta t = 33.5$ fs; (c) $\lambda = 348$ nm, $\Delta\lambda = 8.1$ nm, $\Delta\tau = 17.8$ fs, $\Delta t = 29$ fs and (d) $\lambda = 410$ nm, $\Delta\lambda = 11.8$ nm, $\Delta\tau = 19.5$ fs, $\Delta t = 34.9$ fs.

5. Conclusions

We show that the nonlinear frequency conversion via SFM of ultrashort tunable visible pulses from a NOPA is best performed when the pulses are deliberately left chirped. The mixing with stretched pulses from the Ti:sapphire pump system leads to 20 to 30-fs UV pulses with energies of several μJ and a broad tuning range of 295 - 450 nm. The residual gap around 400 nm can be closed by using a two-stage NOPA with intermediate continuum generation [2]. No compressor is needed in the visible and therefore tuning of the UV wavelength is easy and requires only tuning of the NOPA and a fine tuning of the temporal overlap in the SFM crystal. A special UV prism compressor design is presented, which avoids the need for broadband polarization-rotation elements. We believe that these UV pulses will be of enormous use for ultrafast spectroscopy and for further conversion to even shorter wavelengths.

Acknowledgments