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Generation of 8-fs pulses at 390 nm

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Abstract. We demonstrated the generation of pulses as short as approximately 8 fs at 390 nm, with an average power of 80 mW by frequency-doubling the output of a sub-10-fs, 1.2-MW Ti:sapphire oscillator. Cross-correlation technique was employed to measure the pulse duration. To our knowledge, these are the shortest pulses produced in the violet–blue spectral range at high repetition rates so far.

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With the development of KLM (Kerr-lens mode-locked) Ti:sapphire lasers [1], the generation of ultrashort pulses with durations in the sub-10-fs regime has become possible in the near-infrared wavelength range [2–5]. The advent of dispersion-engineered chirped multilayer dielectric mirrors [6] allowed the construction of femtosecond Ti:sapphire oscillators containing no intracavity components other than the gain medium [7]. This unprecedented simplicity and compactness results in a highly reliable and reproducible performance in the 800-nm wavelength range. However, for many applications, for example experiments on molecules and wide-gap semiconductors, pulses at shorter wavelengths are required.

Generation of higher harmonics and parametric frequency mixing in nonlinear crystals represent powerful techniques for converting pulses from high-repetition-rate oscillators to the blue or even the ultraviolet spectral range. Pulses with a duration of 43 fs centered at 310 nm with a total power of 40 mW by intracavity second-harmonic generation (SHG) of a dye-laser have been reported [8]. More recently, pulses as short as 14 fs centered at 416 nm with a total power of 22 mW have been generated by intracavity SHG of a Ti:sapphire oscillator [9]. The major advantage of using an intracavity doubling scheme is that the power inside a resonator is much higher than in the output-beam. On the other hand, the SHG-crystal has often to be placed at a position in the cavity, where the fundamental pulse is much longer than its minimum duration. Therefore, some of the advantage of high intracavity peak-power is lost by having to double a chirped pulse. Additionally, the frequency of the output beam can not easily be changed back to the infrared region, if required. This can only be done when extracavity doubling schemes are employed. With this technique, the generation of 16-fs pulses at 425 nm with 40 mW average power [10], and 10-fs pulses at 438 nm with 4 mW [11] , respectively, have been reported.

1 Experimental setup

In our experiments we directly doubled the output of a commercially available Ti:sapphire oscillator (Femtosource M1, FemtoLasers GmbH). Pumped with 8 W of a diode-pumped, frequency-doubled Nd:YVO_4 (Spectra-Physics Millenia X), the average output power is 950 mW. The spectrum is centered at a wavelength of 780 nm and has a FWHM of 125 nm, offering the potential to generate 7.5-fs, 1.6-MW pulses at a repetition rate of 75 MHz. The pulses are delivered in a nearly diffraction-limited beam ($M^2 < 1.3$), which turned out to be very important for efficient SHG. To avoid Fabry–Pérot effects, the output coupler of the oscillator is slightly wedged. Therefore a second wedged glass plate has to be placed right after the output to compensate for angular dispersion. This plate can be translated in direction transverse to the beam, so that an adjustable amount of positive dispersion can be introduced into the beam path. Together with some reflections off chirped multilayer mirrors, the phase of the output pulses can be adjusted to some extent.

In contrast to [10, 11] we used reflective optics for focusing the beam into the SHG-crystal as well as for recollimating the generated blue beam. Therefore no prism/grating compressor or chirped mirrors, respectively, have to be used for compensating material dispersion in the path of the blue pulses. A spherical mirror with a radius of curvature of 25 mm focuses the fundamental beam into a 100-μm-thick β-barium borate (BBO) crystal, cut at 29° for type-I phase-matching. The phase-matching condition is fulfilled, when the crystal is slightly tilted with respect to normal incidence of the beam. The tilting is necessary to avoid backreflections into the oscillator which can impair or even stop mode-locked operation. The same optics as for focusing is used for recollimating the beam. Because one mirror folds the beam in

the saggital plane while the other mirror folds it in the tangential plane, both folding angles at the spherical mirrors can be kept small ($\sim 5^\circ - 6^\circ$ full angle) to minimize astigmatism. This results in a blue output with a Gaussian-like circular intensity distribution. The beam-diameter at the focus is calculated to be $\approx 6\,\mu$m with a corresponding confocal parameter of $\approx 70\,\mu$m, which is somewhat shorter than the crystal thickness. This leads to a peak-intensity at the focus of $\approx 8 \times 10^{12}\,\mathrm{W/cm^2}$. Careful optimization of the whole setup yields an average output power of 80 mW in the blue, which corresponds to a conversion efficiency of around 8.5%. The resulting spectrum shown in Fig. 1 (solid line) was recorded by a 1/2 m scanning monochromator and has a FWHM bandwidth of 35 nm. The origin of the sub-structure, which is not present in the fundamental spectrum, is not fully understood yet. The resulting spectrum is much broader than one would normally expect for such a relatively thick doubling crystal.

If the electric field of the fundamental pulses is treated as a plane wave,

$$E(z,t) = A(t - k_\mathrm{f}' z)\exp[-\mathrm{i}(\omega_0 t - k_\mathrm{f} z)] + \mathrm{c.c.}\,, \tag{1}$$

where the carrier frequency ω_0 is, by definition, the first moment of the spectral intensity distribution of the wavepacket, k_f is the wave vector of the fundamental wave (the prime denotes the derivative with respect to ω) and $A(t - k_\mathrm{f}' z)$ is the complex envelope of the pulse, the frequency-doubled spectrum is given by [12, 13]:

$$S(\omega,\tau) \propto \frac{|\chi^{(2)}|^2\omega^2}{n_\mathrm{s}^2}\mathrm{sinc}^2\left\{[\Delta k_0 + (\omega - 2\omega_0)\Delta k_1]\frac{L}{2}\right\} \times \left|\int_{-\infty}^{\infty} \mathrm{d}\omega' A(\omega - \omega')A(\omega')\mathrm{e}^{\mathrm{i}\omega'\tau}\right|^2, \tag{2}$$

where n_s is the refractive index for the second-harmonic wave at $2\omega_0$, k_s is the wave vector of the SH wave, $\Delta k_0 = 2k_\mathrm{f}(\omega_0) - k_\mathrm{s}(2\omega_0)$, $\Delta k_1 = k_\mathrm{f}'(\omega_0) - k_\mathrm{s}'(2\omega_0)$, L is the length of the nonlinear medium, and $A(\omega)$ is the Fourier transform of the complex envelope. In lowest order, a limited phase-matching bandwidth appears due to group velocity mismatch (GVM) between fundamental and second-harmonic waves, restricting the generated bandwidth at the second-harmonic wavelength. As revealed by Fig. 1, the bandwidth of the generated pulses significantly exceeds the phase-matching bandwith calculated in this manner and approaches the GVM-free limit. This shows, that standard calculations, based on plane-wave interaction can not be used any more if the fundamental beam is focused very strongly. A more detailed analysis of this behaviour will be given in Sect. 3.

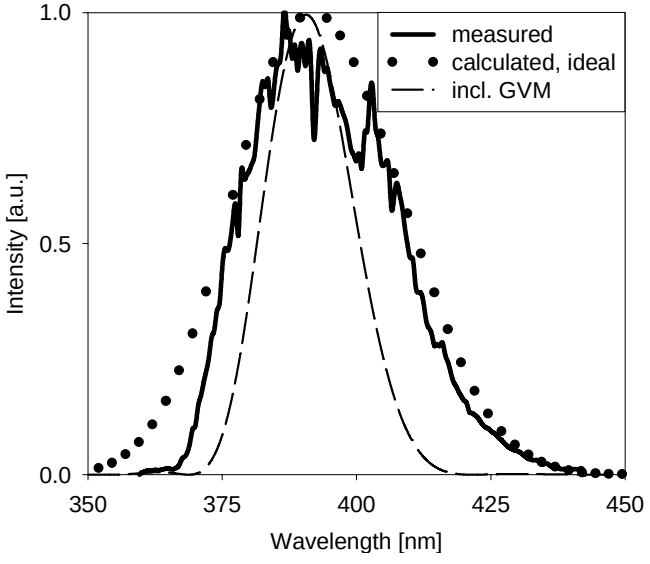

Fig. 1. Measured spectrum of the frequency-doubled pulses

2 Temporal characterization

Standard autocorrelation traces of these violet–blue pulses can not be recorded due to the lack of nonlinear crystals that can be phase-matched for this wavelength range. In BBO, for instance, type-I phase-matching for SHG is only possible down to around 410 nm (type-II phase-matching is frustrated at even longer wavelengths). To avoid these constraints, surface SHG can be applied for measuring ultrashort pulses [14]. But this technique, just as other methods using third-order nonlinearities (for example self-diffraction FROG [15]), would call for pulse energies higher than available directly from an oscillator. Recently, two-photon absorption in GaN photodiodes was used as a nonlinear detector for autocorrelation measurements of fs pulses at 410 nm [16]. But the cut-off wavelength for single-photon absorption of these devices from 365 nm could be critical in our case. Moreover, these diodes are not generally available yet.

To overcome these limitations we measured the cross-correlation function (CCF) between the fundamental and the second-harmonic pulses by sum-frequency generation (SFG). The schematic of our apparatus is shown in Fig. 2. The frequency-doubled pulses co-propagate with the non-converted part of the fundamental pulses behind the doubling-crystal. The two beams enter collinearly the cross-correlator and are separated by the dichroic mirror BS transmitting the fundamental and reflecting the blue pulses (because the shorter wavelengths are more affected by material dispersion). The substrate dispersion is compensated by the chirped multilayer mirror M1. The two beams are focused into

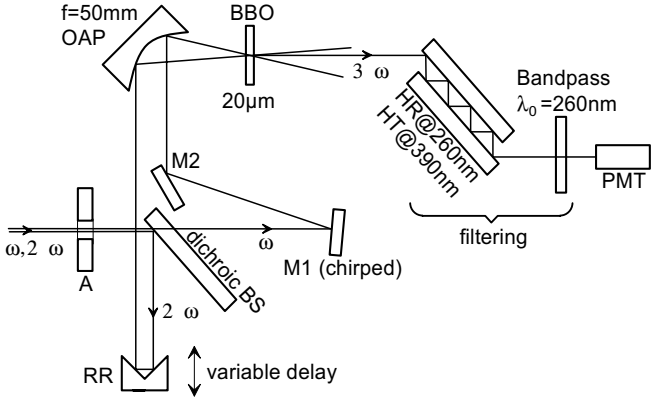

Fig. 2. Schematic of the cross-correlator. A: aperture; BS: beamsplitter; M1: chirped mirror; M2: standard high-reflector (HR790 nm); RR: retroreflector; OAP: off-axis-parabolic mirror; PMT: photomultiplier-tube

a 20-μm-thick BBO cut at 57° for type-II phase-matching for sum-frequency generation (SFG) 780 nm + 390 nm → 260 nm. The crossing angle of the two beams is kept small (smaller than sketched in Fig. 2) to avoid time smearing effects. Since the used photomultiplier tube (PMT) is much more sensitive in the blue than in the ultraviolet regime, careful spectral filtering of the UV beam is necessary. This is done by multiple reflections off dichroic mirrors (M3, M4) which are transparent at 390 nm and high-reflective at 260 nm as well as by a spectral band-pass filter (SF) mounted at the PMT aperture.

The result of the measurement is shown in Fig. 3. In the case of weak focusing into the SFG-crystal and neglecting measurement error due to group-velocity mismatch between the incident waves and the generated UV wave, the cross-correlation signal can be written as [8, 17]

$$I_{\mathrm{CC}}(\tau) \propto \int_{-\infty}^{\infty} \mathrm{d}t\; A^2_{\mathrm{blue}}(t) \times A'^2_{\mathrm{red}}(t-\tau)\,, \tag{3}$$

with

$$A'_{\mathrm{red}}(t) = A_{\mathrm{red}}(t) \otimes \mathrm{sqr}\left(\frac{t}{\Delta k_1\, l_{\mathrm{C}}} + \frac{1}{2}\right)\,. \tag{4}$$

Here $\otimes$ denotes convolution, l_{C} is the length of the nonlinear crystal installed in the cross-correlator and

$$\mathrm{sqr}(x) = \begin{cases} 1 \text{ for } |x| < 1/2 \\ 0 \text{ otherwise.} \end{cases}$$

Neglecting also the group-velocity mismatch between the two incident waves (Δk_1), a simple relation between the FWHM of these pulses (τ_{red} and τ_{blue}) and the FWHM of the measured cross-correlation signal (τ_{CC}) can be found for common pulse shapes [17]:

$$\tau_{\mathrm{CC}} = (\tau^p_{\mathrm{red}} + \tau^p_{\mathrm{blue}})^{1/p}\,, \tag{5}$$

where

$$p = \begin{cases} 2 & \text{for Gaussian pulses} \\ 1.615 & \text{for sech}^2\text{pulses.} \end{cases}$$

But even for our very thin SFG-crystal, it turned out, that Δk_1 can not be neglected for pulse durations in the order of 10 fs. We first measured the duration of the red pulses with a standard fringe-resolved autocorrelator, at the same place, where the cross-correlator was subsequently placed. We determined a FWHM-pulse duration of 10 fs, assuming a sech^2 envelope [18]. Calculating $A'_{\mathrm{red}}(t)$ (4) results in a sech^2-like pulse with a FWHM-duration of 13.2 fs. This corrected value has to be inserted into (5) to get a more realistic estimation of the blue pulse-duration. Finally we measured the cross-correlation function (CCF), and determined a FWMH of 16.7 fs. From these two measurements, a blue pulse-duration of 8.3 fs can be derived. Figure 3 shows the calculated CCF ((3), 20-μm-thick BBO crystal) between two sech^2 pulses of 10 fs (near-infrared, τ_{red}) and 8.3 fs (blue, τ_{blue}), respectively, which appears to be in good agreement with the measured cross-correlation trace.

Doing the same fitting procedure assuming a Gaussian envelope for the SH-pulse results in a blue pulse-duration of 9 fs, but gives rise to an increased discrepancy in the wings between the measured and calculated CCF, respectively. On the other hand, the Fourier-limited pulse-duration, derived from the measured spectrum (Fig. 1), is 7.5 fs. Based on this analysis, we estimate our pulse duration as 8.3 fs ± 0.4 fs.

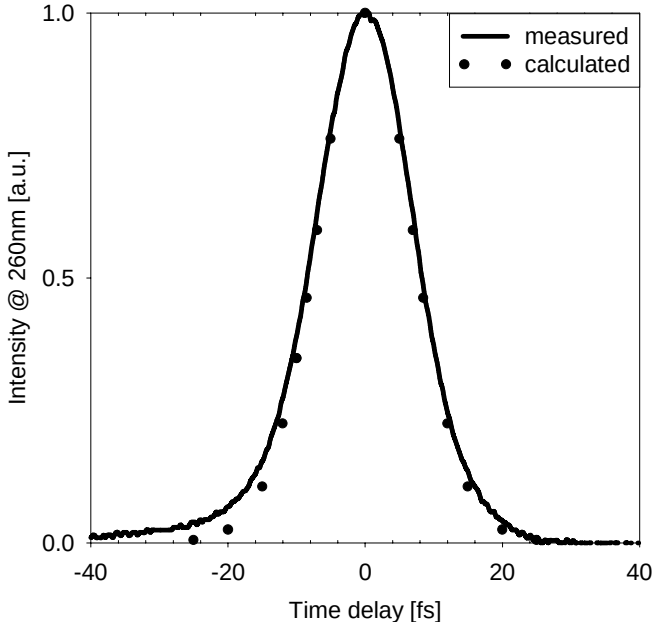

Fig. 3. Measured cross-correlation function (CCF). The *dots* represent a calculated CCF for two sech^2-pulses with 10 fs and 8.3 fs FWHM-duration, respectively

3 Analysis and discussion

The fundamental limitation in ultrashort-pulse SHG is material dispersion. With angle-phase-matching one can achieve that the effective refractive index seen by the fundamental beam (n_{red}) is equal to that seen by the frequency-doubled beam (n_{blue}) at the center frequency, but the refractive index is not constant in frequency. This has two drawbacks: a limited phase-matching bandwidth (PMB) on the one hand, and pulse broadening due to intrapulse group-velocity dispersion (IGVD) on the other hand.

3.1 Phase-matching bandwidth

Calculations, regarding the PMB of SHG-crystals, usually treat the involved light fields as plane waves (like (2)). In the case of very strong focusing, this approximation is not valid any more, mainly as a consequence of two effects, both of them increasing the effective PMB.

First, if the confocal-parameter of the beam becomes comparable to or even shorter than the crystal thickness, second-harmonic radiation will not be generated uniformly over the entire crystal, leading to an effective thickness that is somewhat shorter than the physical thickness, resulting in an increased bandwidth at the expense of a reduced efficiency [12, 19].

The second effect, connected to strong focusing, is achromatic phase-matching. It is shown in [20], that both, phase-matching and group-velocity-matching, can be achieved simultaneously, by using an angular-dispersed fundamental beam. As observed by Ashworth et al. [10], this effect plays an important role, even when the frequency components of the fundamental beam are not separated at the crystal surface. In this case, the SHG-process can be significantly enhanced by those parts of the beam, which are "accidentally" phase-matched, resulting in an enhanced PMB.

3.2 Intrapulse group-velocity mismatch

Transparent optical materials are generally more dispersive for short-wavelength radiation. For our type-I phase-matched BBO-crystal the group-velocity dispersion at 800 nm is 75 fs^2/mm, whereas the frequency-doubled beam sees nearly a three times higher GVD of 201 fs^2/mm. Assuming the ideal case, SHG of fundamental pulses of 10 fs duration will result in blue pulses of 7 fs duration, which would be lengthened in a 100-μm BBO crystal by pure IGVD to 10.6 fs.

Sidick et al. [21] have shown that if the fundamental pulse is not ideally transform-limited and the length of the nonlinear crystal is somewhat larger than the so-called pulse-width-preservation length, there is a complicated interplay between group-velocity mismatch and intrapulse group-velocity dispersion. It is shown in [21], that a quadratic phase of a Gaussian-like pulse maps over to the frequency-doubled pulse, if IGVD is neglected. That means, that if the input beam is negatively chirped, the blue pulses will also be negatively chirped, and IGVD, when taken into account, will therefore tend to shorten the pulses instead of stretching them. We therefore obtained the best results regarding the blue pulse-duration when we used slightly negatively pre-chirped fundamental pulses.

4 Conclusions

We have demonstrated the generation of pulses as short as 8 fs, centered at 390 nm by direct frequency-doubling of the output of a commercially available Ti:sapphire oscillator. By using only reflective optics and carefully controlling the phase of the fundamental beam, no compressor for the blue pulses was necessary. Strong focusing of the fundamental pulses resulted in an increased phase-matching bandwidth. The pulse duration was measured by a cross-correlation technique.

Comparing our results with the performance of previously reported frequency-doubled fs Ti:sapphire oscillators, we may conclude that approximately a factor of two shorter pulse durations and a factor of four higher peak powers could be demonstrated in our experiments. These 0.1-MW-peak-power sub-10-fs blue pulses are expected to benefit a wide range of applications in ultrafast spectroscopy.

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