Deep-UV picosecond flat-top pulses by chirp-matched sum frequency generation

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Picosecond, flat-top, deep-UV pulses are needed to generate high-brightness electron beams to efficiently drive X-ray Free Electron Lasers. Current metal photocathodes have low efficiency and therefore require high-energy pulses, and the generation of high-energy, flat-top pulses in the deep-UV is still challenging. The low efficiencies of both the harmonic generation and the deep-UV pulse shapers restrict the accessible pulse energy. Moreover, the acceptance bandwidth of the harmonic generation limits the minimum rise time of the flat-top profile. In this paper we present the generation of few hundred μJ, picosecond, deep-UV pulses using chirp matched sum frequency generation. This scheme combined with IR spectral manipulation is a novel approach for deep-UV pulse shaping. It permits flat-top pulses with high-energy and fast rise time, highly suited for high-brightness photoelectron beam production.
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High resolution deep-UV (DUV) pulse shaping is required for several experimental fields such as the study of ultrafast phenomena, chemical reactions and atomic quantum states [1, 2]. Here we report the generation of temporally shaped laser pulses to drive high-brightness electron guns. Flat-top, picosecond laser pulses with steep edges can effectively increase the electron beam brightness and consequently enhance the performance of X-ray Free Electron Lasers (FEL) [3, 4]. Current metal photocathodes have low quantum efficiency and require hundreds of μJ at wavelengths between 260-280 nm to provide a charge up to 1 nC.

DUV wavelengths are generated from a Ti:sapphire (Ti:Sa) laser by cascading second harmonic (SH) and sum frequency generation (SFG) between the laser fundamental and second harmonic (ω+2ω=3ω). The pulse shaping can be done prior to or after the harmonic generation. In the first case, the picosecond flat-top pulse is up-converted to DUV, while in the second case a femtosecond pulse can be used in the harmonic generation and then shaped in the DUV. Femtosecond pulses permit efficient harmonic generation, but with this approach the pulse stretching and shaping have to be carried out in the DUV, where such manipulations are rather inefficient. For example, flat-top pulses of only few tens of μJ were demonstrated by using of a UV Dazzler pulse shaper [5].

Shaping chirped pulses in the frequency domain is a flexible and efficient method for shaping IR pulses, but the bandwidth of the chirped pulse makes ordinary harmonic generation difficult. The use of chirp matched (CM) input pulses has been proposed as a solution to overcome the limited acceptance bandwidth of standard harmonic generation [6]. In this letter, for the first time, CM SFG is combined with spectral shaping in the IR to generate shaped high-energy DUV pulses. The output pulses from CM SFG are also chirped, and they can be compressed if desired. Coherent pulse stacking [7] is an interesting alternative for shaping in the IR, but it has lower throughput than a Dazzler. In comparison with passive shaping in the DUV the technique presented here can produce larger output bandwidth and steeper edges [8, 9]. DUV spatial light modulators in 4-f setup [10] can potentially be applied for flat-top shaping, but operation with high energy pulses has not been reported yet.

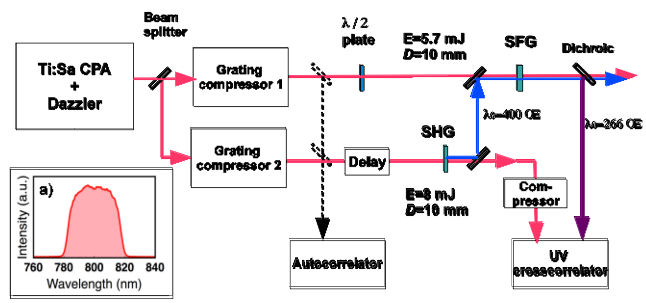

Fig. 1 Sketch of the experimental set up. The SHG and the SFG are BBO crystals cut at 29.2 deg at 44.3 deg respectively. In the inset a) the fundamental spectrum is reported.

CM SFG in nonlinear crystals is based on tailoring the chirp of the input pulses so that at any time the two instantaneous input frequencies produce a perfectly phase matched sum frequency. If the temporal rate of chirp is small, the temporal walk off will be negligible and the above condition can be satisfied along an extended interaction length. A relative long nonlinear medium can then be used to achieve efficient conversion. If both input pulses are derived from the same source, it may be impossible to achieve full temporal overlap because the CM condition constrains the ratio of the pulse lengths. However, for the parts that do overlap the spectral phase

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and intensity of the SFG pulse are simply related to the corresponding properties at ω and 2ω. IR amplitude shaping can be then employed in order to produce the desired spectral intensity in the DUV. CM SFG permits a broad output spectrum because it is not limited by the usual acceptance bandwidth.

Fig. 1 shows the experimental setup. The source is a chirped pulse amplified Ti:Sa laser equipped with Mazzler™ and Dazzler™ acousto-optic shapers. The system can deliver pulses at 100 Hz with variable central wavelength and spectral width [11]. For the present experiments we used 35 nm broad spectra centered at 800 nm, as shown in the inset a) of Fig. 1. The amplified pulses are split and separately compressed, so that the two outputs of 8 and 5.7 mJ, can be independently chirped to fulfill the CM condition. For the considered SFG process, 800(o)+400(o)→ 266(e) nm, the CM condition is satisfied when the group delay dispersion of the fundamental, Φ_1'', and of the second harmonic Φ_2'' are related by $\Phi_1''/\Phi_2'' = -1.79$, as calculated using equations 9 in ref. 6. This ratio can of course be satisfied for different pulse lengths. Our simulations indicate that, assuming fixed fluence, the efficiency of CM SFG can be maintained over a wide range of pulse durations provided that the crystal length is optimized for each duration. We have chosen to work with approximately 5 ps long pulses, which are suitable for our application. For both arms, the beam profile is approximately Gaussian with 1/e^2 diameter D of 10 mm. The energy stability over 500 shots is 0.4 % rms and 2.7% ptp. For both the SHG and SFG 12 mm diameter type I BBO are used. The SHG, which does not take advantage of the chirp matching, is done in short crystals (0.1 or 0.2 mm) to support the necessary bandwidth. The length of the SFG crystal was 0.8 mm, which is close to the optimal value from the simulations. The input IR pulse length is measured with a commercial auto-correlator (APE GmbH). The residual IR pulse from the SHG is recompressed down to 160 fs FWHM and used as gate for DUV cross-correlation, see Fig. 1.

Table 1. Parameters for CU and CM SFG.

Wavelengths			CU	CM
IR pulse λ_0=800 nm	Bandwidth	[nm]	35	
	Duration	[ps]	5.1	
	Chirp Φ_1''	[fs^2]	+4.3·10^4	-4.3·10^4
	Energy E_1	[mJ]	5.7	
SH pulse λ_0=400 nm	Bandwidth	[nm]	13	
	Duration	[ps]	4.9	
	Chirp Φ_2''	[fs^2]	+2.4·10^4	
	Energy E_2	[μJ]	225	
DUV pulse λ_0=266 nm	Duration	[ps]	0.2	3
	Bandwidth	[nm]	0.5	2.5
	Energy E_3	[μJ]	35	220

Numerical simulations are carried out to benchmark the experimental results. The code is based on the coupled equations for the amplitudes in the spatial and temporal frequency domain. Only the spectral amplitudes are restricted to vary slowly compared to the optical wavelengths. Dispersion and birefringence are included [12, 13]. For the simulation, the 800 nm input pulses were calculated by taking the measured spectrum and applying linear chirps which were calculated from the IR autocorrelation measurements and double checked against the compressor parameters.

In the first experiment we compare CM SFG with the case where the input pulses have the same input peak power but are not chirp matched and show that CM SFG yields higher energy and broader bandwidth. The experimental parameters for the three wavelengths, in both the CM and the chirp unmatched (CU) cases, are summarized in Table 1. The only difference between the input beams in the two cases is the sign of the linear chirp for IR pulse.

As reported in Table 1, at the CM condition the available DUV bandwidth and energy are significantly larger than in the CU case.

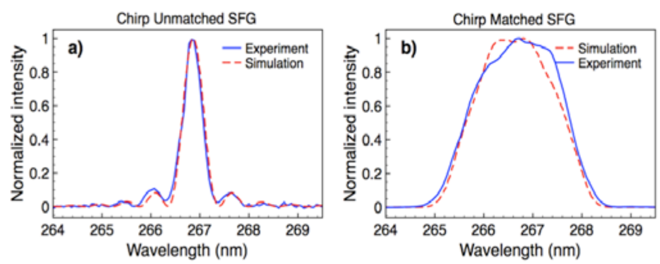

Fig. 2. Simulated (dashed red) and measured (blue) spectra for chirp unmatched a) and chirp matched SFG b).

Figure 2 shows that the measured and simulated DUV spectra are in good agreement. The SFG spectrum for the CU pulses (Fig.2a) approximates a sinc^2 function with 0.5 nm FWHM. In the CM case the spectrum is not limited by the acceptance bandwidth of the SFG and a much broader spectrum of 2.5 nm FWHM could be obtained (Fig.2b). The output energy is more than 6 times larger than in the CU case. It is worth mentioning that in CM case 65% depletion of the second harmonic beam was achieved. The SH energy (E_2 =225 μJ) and its stability (2.8% rms) are the main limitations to the corresponding properties of the DUV beam (E_3 =220 μJ and 2% rms amplitude fluctuation). No degradation of the THG transverse beam profile was observed in the experiment. The simulations indicate that with Gaussian input beams the DUV beam is also nearly Gaussian, with beam quality M^2 <1.05, for both the examples of Fig. 2. The SFG is sensitive to the chirp ratio: for 8% relative variation from the optimal ratio we observed reduction of about 20% in the DUV bandwidth and energy.

In a second experiment we shaped the IR pulses to achieve the flat-top profile required for the X-ray FEL.

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The Dazzler was programmed to produce flat-top spectral intensity, and the laser compressors were set to satisfy chirp matching. Under this condition, the spectral amplitude and phase of the DUV pulse are directly related to the corresponding properties of the IR and second harmonic. The spectral shape through the harmonic generation was not altered by the limited acceptance bandwidth of the SFG. We used an SHG crystal with 0.1 mm thickness to obtain a broad second harmonic spectrum.

At the CM condition, the DUV pulse is linearly chirped with temporal chirp rate equal to the sum of the chirps of the pulses at 800 and 400 nm. When the pulses are long enough that dispersion of the SFG crystal and higher order spectral phase coefficients can be neglected, the temporal intensity approximates the spectral shape [9]. Since our application does not require a transform-limited pulse, we could take advantage of this fact and produce a picosecond flat-top temporal DUV pulse by generating similar shape in the spectral domain. For that purpose the Dazzler filter was optimized to shape a square-like IR spectrum extending over 30 nm with sharp edges of less than 2 nm (10-90% of the maximum intensity).

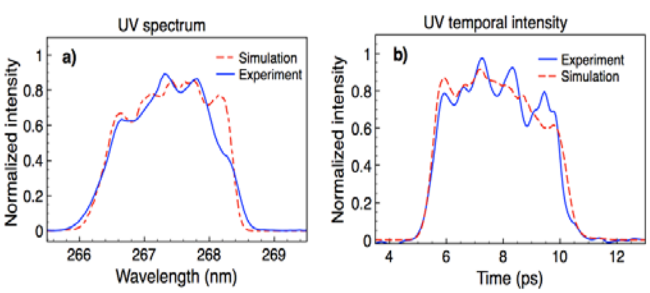

Fig. 3. a) Simulated (red) and experimental (blue) spectra and b) temporal intensity for the DUV flat-top pulse.

Autocorrelation measurements of the IR pulses from compressors 1 and 2 give durations of 6.1 ps and 5.7 ps FWHM, respectively. The group velocity dispersion of the fundamental wavelength and the second harmonic pulses mixed in the SFG were respectively $7.5{\cdot}10^4$ fs^2 and $+4.05{\cdot}10^4$ fs^2.

Fig. 3 shows the DUV spectrum and pulse shape. The simulated curves are in excellent agreement with the experimental profiles. As shown in Fig. 3a), the UV spectrum is flat-top like with more than 2.5 nm width. The edges of the spectrum are quite sharp. The resulting temporal profile measured with DUV cross-correlator is also flat-top Fig. 3b). The pulse duration is 4.5 ps FWHM while rise and fall times (10-90%) are shorter than 0.8 ps and the modulation at the plateau are 10 % rms and 39% ptp. The obtained pulse satisfies the requirements of high-brightness laser-driven electron sources in terms of rise time [4]. Furthermore the achieved energy of 200 uJ permits the generation of sufficient charge for FEL standard metal photocathodes. The measured flat top shape will be tested soon for the generation of low emittance electron bunches.

In conclusion, we demonstrated that the chirp matched sum frequency generation is a feasible technique for the generation of wideband DUV pulses. The broad bandwidth is combined with remarkably high energy for harmonic generation in the picosecond range. We also proved the technique to be highly suitable for efficient high-resolution ps flat-top generation in the DUV, where temporal shaping is still challenging. The shaping tools applied are based on well established technology in the IR. Numerical simulations have been used to accurately reproduce the experimental results in the spectral and temporal domains. In the present setup, the maximum achievable energy and spectral width in the DUV are limited by the SHG. Higher second harmonic energy could be achieved extending the chirp matched scheme to type II SHG. Finally, the presented linearly chirped broadband spectra have potential to be recompressed to achieve intense ultrashort DUV pulses.

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