

Controlling Ultrafast Photoemission via Simultaneous Laser Mixing and Shaping

Randy Lemons^{1,2}, Jack Hirschman¹, Nicole Neveu¹, Joseph Duris¹, Agostino Marinelli¹, Charles Durfee², Sergio Carbajo^{1,3}

¹SLAC National Accelerator Laboratory, Stanford University, 2575 Sand Hill Rd, Menlo Park, CA 94025, USA

²Colorado School of Mines, 1500 Illinois St, Golden, CO 80401, USA

³Department of Electrical and Computer Engineering & Department of Physics and Astronomy, University of California, Los Angeles, CA 90095, USA

*rlemons@slac.stanford.edu

Abstract

We present a novel, versatile framework to generate W-level temporally shaped, near transform-limited, UV picosecond pulses via non-colinear sum frequency generation and demonstrate it producing temporally flat-top, high-power UV pulses capable of enhancing femtosecond- and attosecond-level electron and X-ray free electron lasers brightness. © 2023 The Author(s)

Introduction

X-ray free electron lasers are the brightest x-ray sources currently available and are indispensable to imaging transient atomic and molecular evolution at durations as low as 100s of attoseconds [1]. Their performance – e.g., pulse duration and energy – depends on the quality of electrons produced by RF photoinjectors. The optimal performance of these photoinjectors requires laser pulses, typically in the ultraviolet (UV), with non-Gaussian temporal intensity profiles and durations on the order of 10s of ps [2]. Temporal shaping at these durations is challenging due to the limited spectral content for transform-limited (TL) pulses and a lack of devices to directly modify the temporal amplitude. Pulses of this duration are shorter than the response time possible with currently available electro-optic methods which are used to directly modify temporal amplitude profiles in the nanosecond regime. Alternatively, TL pulses with picosecond duration are too narrowband (<0.1 nm) to be shaped in the spectral domain. Finally, the UV requirement complicates shaping efforts due to possible intensity profile modulations generated during nonlinear conversion and a lack of direct shaping technologies at these wavelengths.

We present a novel framework to simultaneously generate pulses nonlinearly and shape them temporally. Our method consists of a non-colinear sum frequency generation (SFG) scheme where the driving pulses are chirped with equal and opposite amounts of spectral phase that together generate a pulse having a duration and temporal profile roughly equivalent to the combination of the inputs [3]. Since the tailored dispersion relations are imposed on the intrinsically broadband pulses this bypasses the spectral limitations found on TL picosecond pulses. The resulting SFG pulse mimics the spatial quality of the input, is naturally narrowband, and has a flat phase providing resistance to temporal intensity profile distortion during propagation from chromatic dispersion. These characteristics also enable the SFG pulse to be used for further nonlinear conversion without phase distortion. Additionally, as the nonlinear conversion process is fundamentally like optical parametric amplification, it is compatible with high-average power, and high pulse energy implementations. Notable applications of this method are creating sawtooth or square-wave-like temporal pulse shapes with variable duration. We demonstrate the capabilities of this method with a numerical and experimental implementation to generate 26 ps flat-top pulses (Fig. 1B) in the ultraviolet from a 1024 nm, 1 MHz, 40 uJ driving laser designed for MHz-rate next-generation XFELs. Pulses shaped in this way have been shown in simulation to reduce the emittance of electrons generated in RF photocathodes by upwards of 25% (Fig. 1D), allowing for up to 25% higher brightness and shorter wavelengths from XFELs [3].

Results

Numerically, we model the SFG process as type-I mixing in β -Barium Borate (BBO) with split-step Fourier propagation and a Runge-Kutta 4 nonlinear solver. Additionally, we define the phase of the input pulses by adjusting the 2nd, φ_2 , and 3rd, φ_3 , order phase. In Fig. 1A) we show the simulated temporal profile of the SFG pulses after mixing. Notably, these pulses display large temporal oscillations (grey) which are attributed to the interference from high-frequency content in the SFG pulse spectrum. To mitigate these fluctuations, which are detrimental to emittance reduction, we implement a 0.5 nm narrowband bandpass spectral filter to reduce the influence of the unwanted spectral content and flatten the profile towards the desired flat-top (blue). Spectral filtering is optional and application specific.

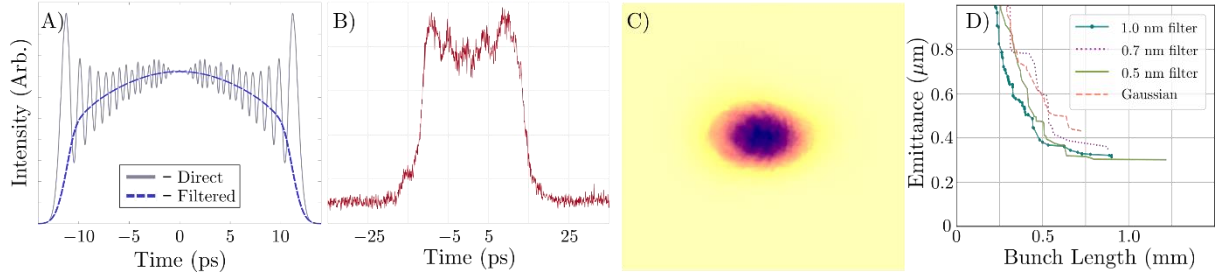


Figure 1: A) Numerically generated temporal profile of the sum-frequency pulse before applying a narrowband filter (grey) and after (blue). B) Experimental temporal profile at 256 nm collected with cross-correlator with 70 fs, 1030 nm oscillator C) 256 nm spatial profile with an ellipticity of 0.64 D) Simulated emittance comparison between temporal Gaussian pulses and shaped pulses with 3 different spectral filters demonstrating improved emittance at all electron bunch lengths

Our experimental implementation of this process is driven by a commercial Yb:KGW laser producing 256 fs pulses centered at 1024 nm with 40 W of power at configurable repetition rates from 100 kHz to 1 MHz. To achieve the pulse stretching and shaping necessary, we require the input pulses to have φ_2 and φ_3 equal to ± 2.561 ps² and ± 0.28 ps³, respectively. This amount of φ_3 is significant and unlikely to be realized in a single device or dispersive element simultaneously with φ_2 . Our implementation thus generates an appropriate amount of φ_3 and corrects φ_2 to be the proper value after. To this end, we split the pulse from the driving laser and pass the copies through a matched grating stretcher and compressor. The stretcher/compressor gratings are 1600 lines/mm reflection gratings which are angled at 50.69° with a grating separation of 338 mm. This results in each pulse having ± 27.31 ps² φ_2 and ± 0.28 ps³ φ_3 . To correct the φ_2 we pass the pulses through opposing ends of a chirped volume Bragg grating (CVBG) designed to add or subtract 24.75 ps² φ_2 with a negligible contribution of high orders of phase. After the CVBG, the pulses are properly conditioned with the required amounts of φ_2 and φ_3 and can then be used in the non-collinear mixing scheme.

Since the harmonic generation is in a non-collinear geometry, the crossing angle between the input beams has a significant impact on the conversion efficiency to the desired SFG beam and not to the parasitic second harmonic generation beams [4]. However, even with this limitation, we were able to realize upwards of 40% conversion efficiency from 1024 nm to 512 nm. Due to the crossing angle, the SFG pulse spatial profile mimics the elliptical overlapping profile of the two incident beams in the crystal rather than simply their spatial profiles. In Fig. 1C) demonstrates the resulting profile of SFG beam being used directly for further nonlinear conversion to 256 nm with an exacerbated ellipticity of 0.64. However, this can easily be corrected with a cylindrical lens telescope; in much the same way, the astigmatism of laser diodes is corrected. Fig. 1B) displays the achieved temporal intensity profile of the 256 nm beam measured with the 70 fs, 1030 nm oscillator pulses in an intensity cross-correlator. This profile is 26 ps FWHM and is characterized by a flattened intense region and faster rise/fall times compared to the Gaussian profile generated without phase addition. The 5 ps oscillations present on the plateau are likely the result of over-filtering in the second nonlinear crystal due to limited acceptance bandwidth and should be easily correctable.

In conclusion, we introduce a framework to simultaneously shape and nonlinearly convert pulses with picosecond duration from femtosecond lasers circumventing the obstacles to applying traditional temporal shaping methods. We experimentally demonstrate this framework by generating 26 ps picosecond UV pulses with temporally flattened profiles, flat phase, and near TL spectral bandwidth at high nonlinear conversion efficiencies. In simulation, pulses shaped in this way have been shown to reduce the emittance of electrons generated in RF photocathodes by upwards of 25%, enabling the next generation of MHz-rate hard x-ray XFELs.

References

- [1] Duris, Joseph et al. (2020). *Nature Photonics* 14.1: 30-36. [2] Musumeci, Pietro et al. (2008). *Physical Review Letters*, 100(24), 244801. [3] Lemons, Randy et al. (2022). *Physical Review Accelerators and Beams*, 25(1), 013401. [4] Kuzmin, Igor et al. (2021) *Applied Optics* 60, 3128–3135.