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Optical Shaping of X-ray Free-Electron Lasers

A. Marinelli,[1,*] R. Coffee,[1,2,†] S. Vetter,[1] P. Hering,[1] G. N. West,[3] S. Gilevich,[1]
A. A. Lutman,[1] S. Li,[1] T. Maxwell,[1] J. Galayda,[1] A. Fry,[1,2] and Z. Huang[1]

[1]*SLAC National Accelerator Laboratory,2575 Sand Hill Road, Menlo Park, California 94025, USA*
[2]*The PULSE Institute, 2575 Sand Hill Road, Menlo Park, California 94025, USA*
[3]*Rose-Hulman Institute of Technology, 5500 Wabash Ave., Terre Haute, Indiana 47803, USA*

In this Letter we report the experimental demonstration of a new temporal shaping technique for X-ray free-electron lasers (FELs). This technique is based on the use of a spectrally shaped infrared (IR) laser and allows optical control of the x-ray generation process. By accurately manipulating the spectral amplitude and phase of the IR laser, we can selectively modify the electron bunch longitudinal emittance thus controlling the duration of the resulting X-ray pulse down to the femtosecond time-scale. Unlike other methods currently in use, optical shaping is directly applicable to the next generation of high-average power X-ray FELs such as the Linac Coherent Light Source-II or the European X-FEL, and it enables pulse shaping of FELs at the highest repetition rates. Furthermore, this laser-shaping technique paves the way for flexible tailoring of complex multi-color FEL pulse patterns required for non-linear multi-dimensional x-ray spectroscopy as well as novel multicolor diffraction imaging schemes.

The X-ray free-electron laser (XFEL) has ushered in a new era for X-ray photon science, increasing by several orders of magnitude the X-ray spectral brightness [1–4]. XFELs now share many of the properties of conventional ultrafast optical lasers such as high-intensity, narrow bandwidth and transverse coherence [5]. One important trait of XFELs is their flexibility, which allows one to control of the spectral and temporal properties of the X-rays by tailoring the electron beam [6–8], the initial seed [3, 9], the undulator [10–12] or by using optical elements during the amplification process [13–15]. The X-ray FEL is fully tunable over many octaves of central fundamental frequency and can be tailored in duration from the few fs level to hundreds of fs.

Control of the pulse duration at the fs time-scale is a key feature for experiments using XFELs. For example, in serial femtosecond crystallography, the intense X-ray pulses from an XFEL can severely damage the sample, thus changing the structural properties of the molecules, destroying the crystalline order within a few tens of fs [16–19]. Generating pulses shorter than the typical damage processes is crucial for the acquisition of high-quality diffraction data for structural biology. Moreover, in time-resolved X-ray spectroscopy, the molecular dynamics initiated by an ultra-fast pump pulse can occur at the tens of fs timescale. Therefore, the experimental investigation of these phenomena require X-ray pulses of sub-10 fs duration [20, 21]. Furthermore, the ability to extend the nonlinear optical techniques used in ultrafast chemistry and atomic and molecular physics to the x-ray regime is viewed as a cornerstone for the coming decade of Free Electron Laser development [21].

The LCLS produces typical x-ray pulses that vary from ∼100 fs at soft x-ray wavelengths to 25 fs for hard x-ray wavelengths when the accelerator operates in a standard mode of 180 pC electron bunch charge with nominal electron bunch compression. One possible option to achieve shorter pulse durations is to operate at a lower electron bunch charge, e.g. a 20 pC, which typically produces 15-20 fs duration pulses at soft X-rays and 5-10 fs at hard X-rays. A more flexible and real-time tunable alternative that allows active control of the pulse duration is based on a nominal bunch charge but selectively suppressing the lasing action for all but a small fraction of the electron bunch longitudinal dimension. This allows the FEL instability to develop only over a fraction of the electron bunch, thus resulting in a shorter x-ray pulse output. This alternative is typically accomplished using a thin slotted aluminum foil placed in a high-dispersion region of the accelerator called the emittance spoiler [7, 22]. Compared to the low-charge operation, selective emittance spoiling generates a small background, typically below 0.1% of the saturation power at x-ray energies. Successful under most operating conditions, this method of x-ray pulse duration control becomes less favorable at very low electron bunch energies, which produce photon energies below the 540 eV Oxygen *K*-edge at LCLS.

Short pulse tailoring for high-average power FELs is an important subject for the future of X-ray science. The next generation of XFELs will be based on high average power superconducting linacs. For example, the LCLS-II linac will generate an average beam power close to 1 MW [23]. The use of beam shaping techniques based on any solid target electron bunch interaction such as the foil-based emittance spoiler will likely fail for such high average power. However, the generation of short and adjustably shaped x-ray pulses is of particular interest for these next generation machines [21]. The use of lasers to tailor the phase-space of the electron beam is particularly advantageous in this context, since high power lasers can be stably operated at MHz repetition rates. Use of IR lasers to manipulate the properties of high-gain FELs has previously been reported in the context of multicolor

*Electronic address: marinelli@slac.stanford.edu
†Electronic address: coffee@slac.stanford.edu

seeded FELs [24], as well as seeded microbunching instability experiments [25].

We report here the demonstration of a new laser-based scheme for x-ray pulse shaping at the fs time-scale. The experiment was performed at the Linac Coherent Light Source of the SLAC National Accelerator Laboratory. The experimental setup is illustrated in Fig. 1. The X-ray pulse duration is controlled by tailoring the longitudinal phase-space of the electrons with a temporally shaped laser heater. The laser heater is placed at the exit of the LCLS photo-injector where it is normally used to slightly increase the energy spread of the electron beam to suppress collective beam instabilities that develop along the accelerator [26, 27]. This is accomplished by resonant interaction of the electrons with an infrared (IR) laser in a magnetic undulator. Since the FEL process is especially sensitive to the electron beam energy spread [28], one can suppress the lasing altogether by increasing the energy spread beyond the lasing threshold (typically $\sim 0.1\%$ at X-ray energies). By temporally shaping the laser heater pulse, for example generating a temporal notch, we can selectively suppress the FEL instability along unwanted parts of the electron bunch and thus tailor the duration of the resulting X-ray pulses.

A key feature of this laser heater pulse shaping scheme is that the phase-space manipulation happens before the bunch-compression process. This means that the duration of the x-ray pulse is given by the duration of the temporal notch in the heater pulse divided by the compression factor. In a typical XFEL the compression factor is on the order of 100. This means that the x-rays can be controlled at the fs time scale by shaping the IR laser pulse at the hundreds of fs timescale, well within the capability of commercially available fs laser systems. More generally, the temporal resolution of the shaping process is given by:

$$\tau_x \simeq \frac{\tau_{IR}}{C}, \tag{1}$$

where τ_{IR} is the Fourier-limited duration of the IR pulse (in field amplitude) and C is the compression factor.

In our experiment, the heater is a Ti:Sapphire laser with a central wavelength of 760 nm and a bandwidth of 3.5 nm FWHM, with a corresponding Fourier-limited duration of roughly 340 fs in field amplitude (240 fs in intensity). The IR laser was shaped with an acousto-optic programmable dispersive filter (AOPDF) [29, 30], placed at the exit of the IR oscillator. The AOPDF is a DAZZLER, manufactured by Fastlite. The laser pulse was stretched and amplified through a regenerative amplifier followed by a multipass amplifier and finally partially re-compressed. To cover the entire length of the electron bunch (roughly 6 ps before compression), the laser heater was only partially re-compressed to a duration of roughly 8 ps.

The electron beam is accelerated to an energy of 5.8 GeV. The corresponding photon energy emitted in the LCLS undulator [1] is roughly 1.5 keV. The longitudinal phase-space of the electrons can be measured at the end of the undulator beamline by means of an x-band deflecting cavity as described in [31]. Figure 2 shows the measured longitudinal phase-space of the electron bunch at the end of the undulator line for an unshaped electron bunch, as well as a longitudinally shaped bunch. To reconstruct the properties of the X-rays, the data was measured with lasing suppressed by a perturbation in the orbit (A and C), as well as with lasing on (B and D). The duration of the compressed bunch is roughly 100 fs FWHM. The bunch charge is 180 pC, with a peak current of 1.5 kA. For the unshaped bunch, the slice energy spread of the core of the beam is roughly 4.5 MeV FWHM (1.9 MeV root mean square (RMS) for a Gaussian distribution). The X-ray temporal profile can be reconstructed by mapping the distribution of the energy loss of the electrons. The unshaped bunch emits an X-ray pulse with a duration of 55 fs FWHM, and a peak power of 35 GW. To control the duration of the X-rays, the energy spread was selectively increased to 9 MeV FWHM, in all of the bunch except for a short lasing core of 10 fs (see Fig. 2(C, E)). This was achieved with a laser heater pulse energy of 300 μJ. The resulting pulse duration in this case is roughly 10 fs FWHM (Fig. 2(F)). The AOPDF can be used to tailor the phase and amplitude of the spectrum of the IR laser. This allows a high degree of flexibility in tailoring the X-ray pulse. Figure 3 shows the X-ray profiles reconstructed from the XTCAV, generated using three different spectral filters, as well as the amplitude and phase of the filters. In the first two cases notch filters with a width of 1.1 nm and 0.5 nm were used. For our partially compressed laser pulses, these correspond to temporal notches of roughly 2 ps and 1ps respectively. After compression, the resulting pulse durations for these shots are 34 fs and 17 fs FWHM respectively. The shortest pulses were achieved with a "phase-flip" filter, which has a constant amplitude and a flat phase switching from 0 to π in the center of the spectrum. For our 20x Fourier limited pulses, this type of filter generates a triangular notch in time with a duration of roughly 1 ps FWHM. When exponentiated through the FEL gain, this yields an X-ray pulse shorter than the temporal notch divided by the compression factor; in fact for such a triangular notch, the half-point of the laser field can still be large enough to suppress the lasing, depending on the maximum laser power. For the case shown in Fig. 3 the pulse duration is 11 fs FWHM.

Figure 4 (A) shows the average FWHM pulse duration for the three filters, as well as the unshaped case. The pulse duration is 52 fs for the unshaped bunch, 40.5 fs for the 1.1 nm notch, 20 fs for the 0.5 nm notch and 12 fs for the phase-flip. The average pulse energy (Fig. 4 (B)) scales roughly proportionally to the pulse duration going from 320 μJ for the phase-flip to 1.8 mJ for the unshaped bunch (the unshaped bunch has a slightly higher peak power than the other three datasets due to a better optimization of the slice energy-spread of the lasing core). From Fig. 4 the fluctuation level on the pulse duration for the shaped pulses is roughly 20%, larger than the un-

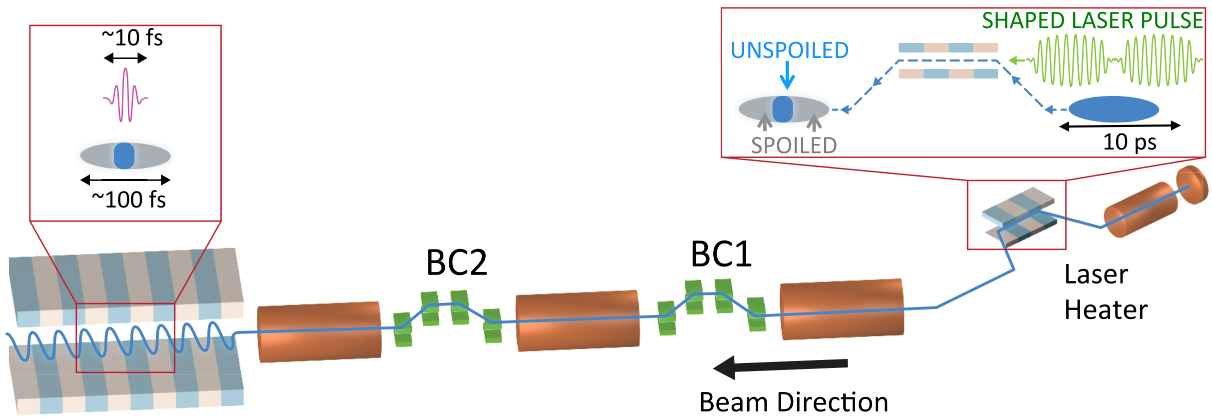

FIG. 1: Schematic representation of the experiment(not to scale). From right to left: an electron bunch is generated with a photo-injector and accelerated and compressed in the LCLS linac. The longitudinal phase-space of the bunch is tailored by means of a temporally shaped IR pulse in the laser heater, leaving a 1 ps-long fraction of the bunch unspoiled. After compression, this unspoiled fraction of the electron bunch emits a short ($\sim$ 10 fs) X-ray pulse in the LCLS undulator.

shaped case. This is due to the residual microbunching instability in the unspoiled part of the bunch, and could be fixed by more accurate tuning of the unspoiled energy spread. The pulse-energy fluctuations are higher for the shorter pulses, consistently with the theory of SASE FELs [32]. To quantify the level of FEL background introduced by the optical shaper we measured the FEL intensity for the same IR pulse energy as the shortest notch case. The measured suppressed FEL pulse energy was roughly 10 μJ.

While in this Letter we report the simple case of a single pulse of variable duration, we note that this general concept allows for near arbitrary shaping of x-ray pulses, with the laser shape being constrained only by the available laser bandwidth. Given the flexibility of IR pulse shaping techniques, this method could potentially be used for generating arbitrary pulse structures, with application to multicolor/multibunch FELs [6]. For example, one could generate multiple pulses of arbitrary duration, overcoming the limitations of the emittance spoiler which required installation of a new foil if a new type of pattern is required. Moreover, with proper optimization of the system to reduce fluctuations in the temporal domain, one could use lock-in amplification style techniques to reduce the impact of noise on the experimentally recorded data. For example, at a high repetition rate FEL, the X-ray profile could be varied from shot-to-shot with a known kHz scale duty-cycle. By mixing the resulting signals with a reference oscillation of the duty-cycle, one can dramatically increase signal fidelity for fundamentally weak processes such as impulsive Raman excitation [33] and multi-color wave-mixing techniques [34] in the x-ray regime. Further, if such controlled modulation can run at a duty-cycle that competes with the dominant sources of noise and jitter in the machine, then this control could be wrapped into a feedback system that stabilizes such parameters as FEL pulse shape, duration, arrival time, etc.

In conclusion, we have demonstrated a laser-based technique to tailor the temporal profile of XFEL pulses at the femtosecond time-scale. The method has similar performance as the currently used technique of emittance spoiling for the same bunch charge yet addresses a broader range of FEL operating conditions than the bulk material spoiling method. We have demonstrated a 5-fold reduction of x-ray pulse duration, generating pulses as short as 11 fs FWHM starting from a 50 fs unshaped X-ray pulse. Unlike other techniques currently in use, this method is directly applicable to high average power machines and paves the way to fs-level shaping for the next generation of XFELs. Furthermore, this method is the first step towards arbitrary shaping of multicolor pulses at the fs timescale and the use of lock-in detection for user experiments at next-generation XFELs.

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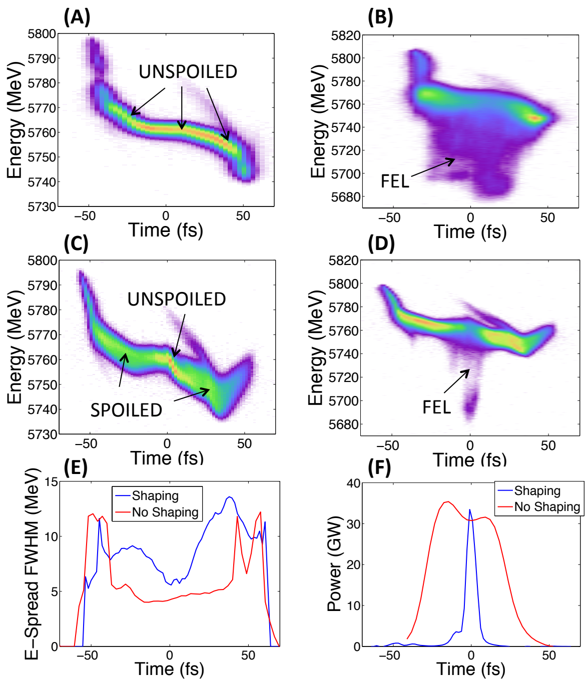

FIG. 2: **(A,B)**Longitudinal phase-space of the electron bunch with lasing suppressed and lasing on, respectively, for the unshaped electron bunch. **(C,D)** Longitudinal phase-space of the electron bunch with lasing suppressed and lasing on, respectively, for the temporally shaped electron bunch. **(E)** Slice energy spread as a function of time for the unshaped electron bunch (red) and the temporally shaped electron bunch (blue). **(F)** X-ray profile for the unshaped electron bunch measured from the energy-loss to the FEL (red) and the shaped electron bunch (blue)

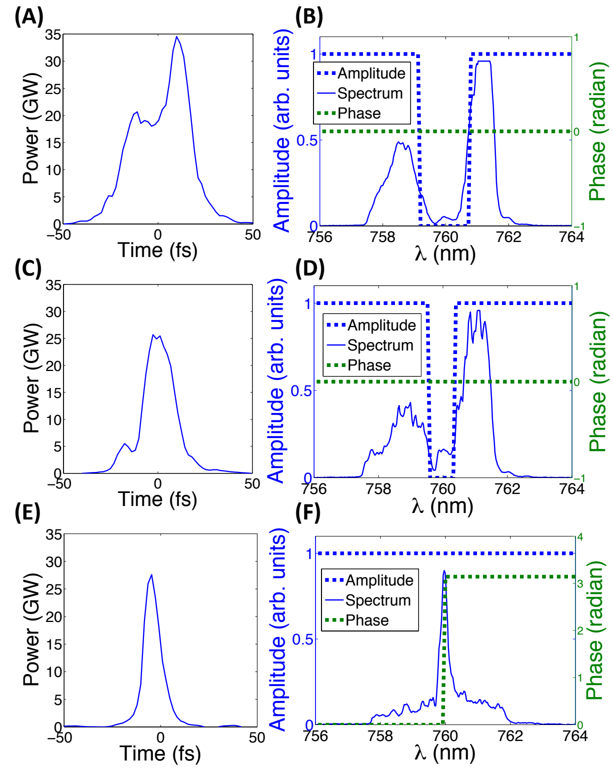

FIG. 3: **Left plots (A,C,E):** X-ray temporal profile for three different IR spectral filters. **Right plots (B,D,F):** corresponding amplitude and phase of the IR spectral filter and spectrum measured after amplification.

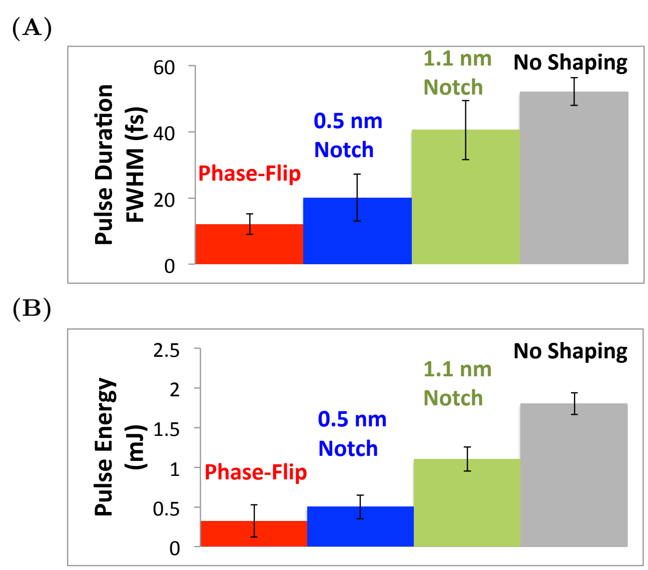

FIG. 4: **A**: average FWHM pulse duration for different IR filters. **B**: average pulse energy for different IR filters.

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