

Demonstration of Mode-Locked Frequency Comb for an X-Ray Free-Electron Laser

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X-ray free-electron lasers (FELs) are powerful photon sources offering a wide wavelength range, subfemtosecond pulse duration, and high brightness. Most x-ray FELs are based on self-amplified spontaneous emission (SASE). SASE-FEL radiation has excellent transverse but only limited longitudinal coherence, with power and spectral profiles consisting of multiple randomly distributed spikes. In this Letter, we present the first experimental demonstration of mode-locked SASE, which generates periodic trains of phase-locked subfemtosecond pulses, thus providing an x-ray analog of the optical frequency comb. Our approach combines the mode-coupled SASE scheme, where magnetic chicane between the undulator modules of the FEL increase the coherence of the output radiation, and an external optical laser that restricts the FEL amplification to periodic and short regions of the electron bunch. The work relies on evidence in the frequency and time domains for photons and electrons, respectively, and will benefit investigations of ultrafast dynamics as well as coherent spectroscopy, and enable new types of experiments requiring phase-correlated x-ray pulses.

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X-ray free-electron lasers (FELs) generate high-power, femtosecond and attosecond, transversely coherent pulses, which are employed for experiments in multiple scientific disciplines, ranging from biology to atomic physics [1,2]. Their high brightness has enabled ground-breaking techniques such as serial femtosecond crystallography and single-particle imaging, advancing our understanding of protein structures, including challenging targets such as *G* protein-coupled receptors [3] and other macromolecules [4–6]. They have also opened new frontiers in atomic, molecular, and optical science, as well as nonequilibrium chemistry and condensed matter physics [7]. The success of

pioneering facilities such as the Linac Coherent Light Source at Stanford has spurred the development of XFELs worldwide [1]. Current research aims to improve the coherence of the XFEL pulses, and eventually to generate arbitrary x-ray waveforms, enabling new applications to many branches of science [8]. XFELs generate radiation from a high-brightness multi-GeV electron beam traveling through an undulator. They commonly depend on the self-amplified spontaneous emission (SASE) mechanism, which builds on the intrinsic shot noise of the electron beam [9,10]. SASE results in x-ray pulses of limited temporal coherence with durations matching the electron bunch length, consisting of multiple uncorrelated spikes in time and frequency.

Different schemes to produce fully coherent (both spatial and temporal) radiation have been put forward. One strategy is to reduce the duration of the electron bunch to the subfemtosecond scale, so that only one SASE spike is present and the output becomes short and temporally coherent [11–17]. Alternatively, there are methods that

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increase the coherence length. Notable examples are seeding schemes, where FEL amplification starts with a coherent input instead of the stochastic shot noise SASE builds on. Seeding has been demonstrated from a filtered SASE signal (self-seeding) for both soft and hard x-rays [18–22], as well as with external optical seed lasers for soft x-rays [23,24].

The temporal coherence can also be improved by delaying the electron beam with respect to the photons using multiple magnetic chicane in the undulator beam line [25–27]. Thereby, the x-ray photon field generated by certain electrons overlaps with that of other electrons after being delayed, and phase correlations between these parts are inherited by subsequent FEL processes. This occurs because the phase information is present both in the radiation field as well as in the electron beam bunching. The chicanes allow for the following schemes: (i) high-brightness SASE (HB-SASE) [26], where chicanes are set to different delays to produce radiation with reduced bandwidth with respect to SASE; (ii) mode-coupled SASE (MC-SASE) [25], where all chicanes have the same delay, resulting in frequency combs; and (iii) mode-locked SASE (ML-SASE) [25], which uses chicanes with the same delay, as in the MC-SASE scheme, but combined with an external optical seed laser with a period matching the chicane delay and slippage per undulator module, limiting the lasing to certain periodic parts of the electron bunch. In the time domain, ML-SASE results in a temporal profile of equally separated and carrier-phase-correlated pulses. In the frequency domain, the periodic gain modulation induced by the seed laser causes the sidebands created by the MC-SASE mechanism to acquire further sidebands. These sidebands overlap with the adjacent frequency modes, resulting in narrower frequency modes and a larger total bandwidth compared to MC-SASE. In short, the delays synthesize a microscale optical cavity traveling at light speed, which gives simultaneous access to pulse durations inaccessible with SASE and photon energies inaccessible with conventional lasers. Following the demonstration of HB- and MC-SASE [27], here we present the experimental verification of the ML-SASE scheme and show evidence in the frequency and time domains. We note that there are also alternative schemes to produce two or more phase-locked pulses with XFELs, for instance, based on ultrashort emission [28] or from other types of seeding [29,30].

Mode-locking techniques at XFELs offer new possibilities of operating these photon sources, yielding a huge variety of outcomes, including trains of attosecond pulses for multichromatic single-shot spectroscopy [31], wide-bandwidth pulses [32], and fully coherent pulses with meV bandwidth [33]. These advances enable time-domain interferometry and coherent control of quantum states [30], and therefore open new avenues for research in physics, chemistry, and biology at angstrom–subfemtosecond scales [1].

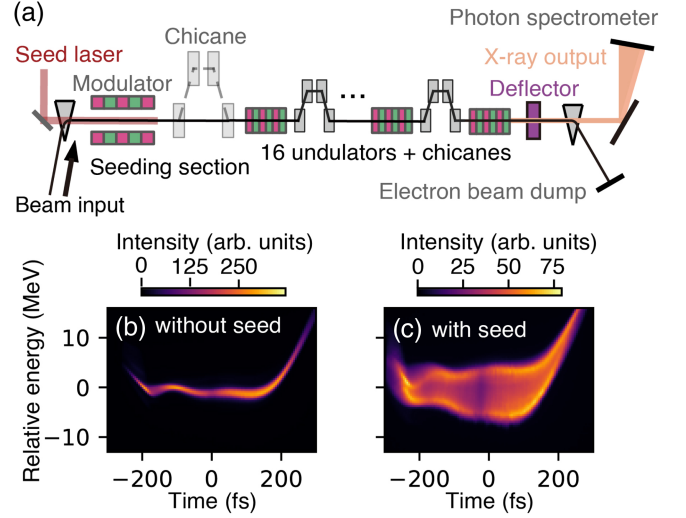


FIG. 1. (a) Schematic layout of the experiment. (b) and (c) Longitudinal phase space measurements of the electron beam without and with the 790-nm seed laser, respectively.

We demonstrate ML-SASE at the Athos soft x-ray beam line of SwissFEL [34] for photon energies between 500 and 600 eV. Figure 1(a) shows a schematic of the experimental layout. The input energy of the electron beam is between 3.2 and 3.4 GeV, the bunch charge is 200 pC, and we employ a helical undulator configuration (corresponding to circularly polarized radiation). There are two seeding sections before the undulators, each of them consisting of a modulator and an optical laser system. Here we use the first seeding section. The modulator has effectively eight 0.2 m-long periods with a maximum K value of 36 [35]. The seed laser [36] is based on a titanium-sapphire system and includes harmonic conversion components capable of delivering optical pulses with 790, 395, or 263.3 nm wavelengths. The full width at half maximum (FWHM) of the pulses can be adjusted between around 60 and 450 fs, where the maximum pulse energy is ≈ 4.5 mJ for a wavelength of 790 nm, and ≈ 0.15 mJ for 263.3 nm. There is a magnetic chicane in the seeding section that can convert the laser-induced energy modulation into an electron density modulation. The lasing restriction in ML-SASE can be achieved by either energy or density modulation [32], but to avoid complexity due to additional compression and related space-charge effects, we restrict ourselves to pure energy modulation, i.e., the seeding chicanes are turned off. Finally, the beam is sent to the undulators for lasing, where the magnetic chicanes build up phase correlation of the output. The undulator line consists of 16 modules, each with 52 periods of 38 mm corresponding to a total length of 2 m, and a maximum K value of 3.8 [37]. There is a chicane placed between every two undulator modules, and each of them can delay the electrons by up to about 5 fs, corresponding to ≈ 1.5 μm .

For electron beam diagnostics, we measure the longitudinal phase space (LPS) with an x-band radio-frequency transverse deflecting structure (TDS) [38,39] and a scintillating screen [40] placed after the undulators. In addition to establishing spatial and temporal overlap between the electron beam and the seed laser, comparison of LPS measurements with lasing-on and -off conditions permit reconstruction of the FEL temporal profile [41,42]. Two LPS measurement configurations are employed: one with lower (few-femtosecond) resolution, which allows us to record the full electron beam on the screen, and another with higher (subfemtosecond) resolution, capable of distinguishing the energy modulation at 790 nm as well as individual lasing spikes.

For photon diagnostics, a monochromator [43] yields pulse-by-pulse spectra. Its energy calibration is verified by photon absorption measurements of the spectral line separation of the nitrogen $1s \rightarrow \pi^*$ transition. In addition, we also use a gas-based x-ray pulse energy monitor [44].

For the ML-SASE demonstration, we first establish and optimize the MC-SASE output [27]. In particular, we operate with a relatively low peak current of a few hundred Amperes, resulting in an FEL gain length comparable to the undulator module length. Correspondingly, the energy spread of the electron beam is relatively low, allowing for large chicane delays without spoiling the bunching of the electron beam. The total bunch length amounts to ≈ 500 fs [see Figs. 1(b) and 1(c)].

Next, we set up the seeding stage for ML-SASE. Initially, we used a 790-nm seed laser with a FWHM duration of ≈ 425 fs and a pulse energy of ≈ 4.2 mJ. Results for the 263.3-nm seed laser obtained at a later stage are presented in End Matter (Appendix A). After establishing a coarse spatial and temporal overlap between the electron beam and the seed laser, we utilize LPS measurements for further optimization of the seed laser timing, the transverse overlap in the modulator between the laser and the electron beam, and the modulator K parameter. With the few-femtosecond resolution of this measurement configuration, we observe an overall increase in the time-resolved beam energy spread, which is proportional to the amplitude of laser-induced energy modulation. For the case shown in Fig. 1, a root mean squared (rms) energy spread increase of more than 4 MeV is observed. The modulated electron beam is then sent to the undulator line for lasing. We operate close to FEL saturation, which requires nine undulator modules for the 790-nm case. The chicane delays are set to 2.1 fs, an empirically optimized value close to the ideal matching condition of 2.2 fs (the seed laser wavelength of 2.63 fs minus the slippage per module of about 0.43 fs) that gave maximum pulse energy. Similar chicane delay settings are used for MC- and ML-SASE to allow for a meaningful comparison between the operating modes.

Figure 2 shows single-shot and averaged spectra of MC- and ML-SASE using the 790-nm seed laser. While both

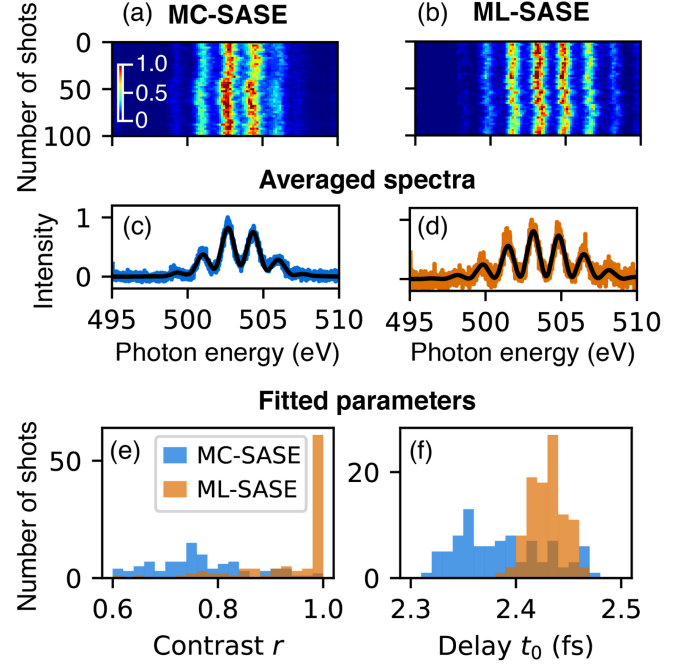


FIG. 2. Spectral measurements. (a) 100 consecutive single-shot spectra measured for MC-SASE with a 790-nm seed laser. (b) Equivalent measurement for ML-SASE. We remove a constant background from the spectra, and normalize them by their average. (c),(d) Averages of the data shown in (a) and (b) along with fits using Eq. (1). (e),(f) Histograms of the fitted contrast r and delay t_0 in Eq. (1) for (a) and (b), respectively.

outputs show reasonably stable frequency combs, there are differences. Intensities are lower for ML-SASE, consistent with the restricted lasing regions: the average pulse energy is ≈ 30 μ J for MC-SASE and ≈ 7 μ J for ML-SASE. The spectral measurements are qualitatively consistent with the simulations of Thompson and McNeil [25]. The ML-SASE spectra, compared to the MC-SASE spectra, show a broader envelope and a greater number of visible modes. The individual modes from single shots are also found to be narrower in Fig. 2. These features all indicate that, as expected, the modulation applied to the electron bunch causes the individual modes to develop sidebands which overlap with, and drive, the neighboring modes, leading to phase locking, a broader total bandwidth and correspondingly shorter pulse duration.

For a quantitative comparison between MC- and ML-SASE at 790 nm, we fit the spectra with a product of a Gaussian envelope and an interference term:

$$P(E) = P_0 e^{-\frac{(E-E_c)^2}{2\sigma^2}} \{1 + r \cos[t_0(E - E_{\text{int}})/\hbar]\}, \quad (1)$$

where P_0 , E_c , σ are the fitted amplitude, central photon energy and rms bandwidth of the envelope, respectively, r is the contrast of the interference pattern, t_0 is the delay that corresponds to the spectral line separations, E_{int} reflects the

change in spectral line positions, and $\hbar$ is the reduced Planck constant. The fit results for the averaged spectra are shown as black lines in Figs. 2(c) and 2(d) [see End Matter (Appendix B) for single-shot examples]. For ML-SASE, we obtain $\sigma = 2.5 \pm 0.1$ eV and $r = 0.94 \pm 0.09$. For comparison, MC-SASE has $\sigma = 1.8 \pm 0.1$ eV and $r = 0.76 \pm 0.10$. We also find that for ML-SASE, the fitted $t_0 = 2.43 \pm 0.02$ fs approximately matches (4% smaller) the summed slippage within one undulator module of 0.43 fs plus the chicane delay of 2.10 fs. The fitted t_0 is about 7% smaller than the seed laser wavelength (790 nm or 2.63 fs), probably due to a frequency chirp of the seed laser or spectrometer error. Also, the histograms in Figs. 2(e) and 2(f) reveal that mode locking very substantially improves the shot-to-shot jitter of the period t_0 and the contrast r , two of the key parameters describing a frequency comb. Quantitatively, the seed laser assures that over half of the single shots have r indistinguishable from 1 and therefore feature strong phase coherence among pulses; without the seed laser, r almost never approached 1. In addition, the seed laser reduced the jitter in t_0 from 60 to 20 as (i.e., from 2.4% relative jitter to 0.8%), representing a threefold improvement from 40 to 13 meV in the shot-to-shot stability of the comb frequency ($2\pi\hbar/t_0 = 1.67$ eV). Both MC- and ML-SASE have similar jitter of the other parameters E_c (0.2 eV) and E_{int} (0.14 eV), which originate from the electron beam energy jitter.

To see easily in the time domain what our analysis using Eq. (1) has already shown, we consider Fourier transforms of the spectra in Figs. 2(a) and 2(b) averaged in two ways: First, we Fourier transform the averaged spectra, which will include energy smearing due to the shot-to-shot phase jitter derived from the electron beam jitter. The amplitudes of these traces, shown in Fig. 3(a), reflect the temporal coherence of the output radiation [45]. We observe spikes at around 2.5 and 5.0 fs for MC- and ML-SASE, respectively, corresponding to the seed laser period and its double and reflecting the increase of the temporal coherence from the undulator chicane. As the chicane settings are equivalent for both schemes, their traces look similar. Second, we average the amplitudes of Fourier-transformed single-shot spectra, a procedure which removes the overall phase jitter accounted for by E_{int} in Eq. (1). These traces, shown in Fig. 3(b), reflect the averaged temporal autocorrelation of the output radiation fields. We observe an overall exponential decrease in intensity of both traces due to the spectrometer resolution limit (here around 0.2 eV or 20 fs). Within this range, ML-SASE shows more than 12 equidistant spikes, while for MC-SASE only a few peaks are visible. This is consistent with ML-SASE producing pulse trains with equal delays of ≈ 2.5 fs, while with MC-SASE the output is a superposition of multiple uncorrelated pulse trains. We have further evidence for ML-SASE by observing the spectral properties when scanning the chicane delay and the seed laser pulse energy [see End Matter (Appendix C)].

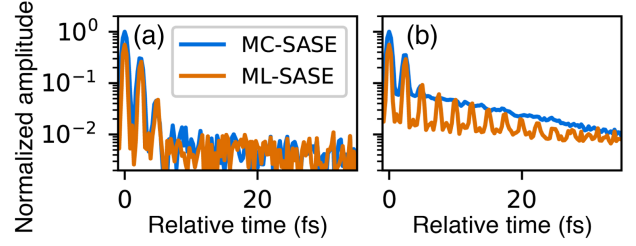


FIG. 3. Fourier analysis of the single-shot spectra in Figs. 2(a) and 2(b) for MC- (blue) and ML-SASE (orange). (a) Amplitudes of the Fourier transform of the averaged spectra, which reflect the temporal coherence of the entire spectrum. (b) Averages of the amplitudes of single-shot Fourier transforms, which reflect the averaged autocorrelation of the output field in the time domain. All data are normalized by the maximum of the corresponding MC-SASE traces.

Shifting from photon spectroscopy in the frequency domain, we focus on time-resolved measurements of ML-SASE using the TDS [39,41,42], performed with the 790-nm seed laser during a consecutive beam time. First, we compare the LPS measurement of the whole electron beam with and without lasing—that is, with optical seed laser on but with and without undulators contributing to the SASE—to estimate the duration of the entire FEL output, found to be ≈ 300 fs. Next, we use the high-resolution LPS configuration to resolve individual lasing pulses. The time calibration is obtained from the known laser field period of the energy modulation, here 2.6 fs. Figures 4(a) and 4(b) show single-shot LPS measurements of the central part of the electron beam. We clearly observe the energy modulation induced by the seed laser for ML-SASE. In addition, one can observe that when turning on the FEL process, the energy spread increases at certain, periodically separated locations within the electron beam (also visible as longitudinal sections with lower intensities), indicating that lasing only occurs at these locations. By contrast, with MC-SASE the increase in energy spread, and therefore also the lasing, proceeds everywhere along the bunch.

For a more quantitative analysis, we fit the central energy $E_c(t)$ using the sum of a sine and a linear function:

$$E_c(t) = A \sin(\omega t + \phi_0) + Bt + C, \quad (2)$$

where A , ω , ϕ_0 , B , and C are fitting parameters. The fit results for the data in Figs. 4(a) and 4(b) are shown as black lines. Vertical slices of the measurements can be assigned a relative phase $\phi_{\text{fit}}(t) = \omega t + \phi_0$ with respect to the energy modulation. Figure 4(c) shows the time-resolved energy spread for lasing-on and -off conditions after adjusting the relative timing between the two conditions. For lasing-off conditions the reconstructed energy spread shows a sinusoidal behavior, reflecting the effect of the slice mean energy variation being smeared out over a time range given by

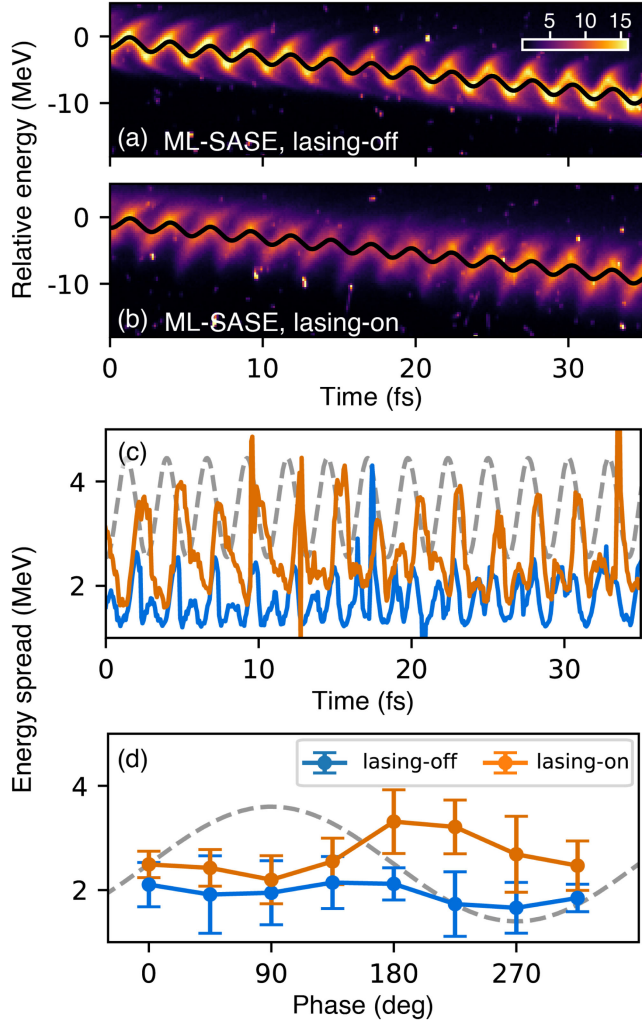


FIG. 4. Time-resolved measurements and analysis of two single shots obtained with the 790-nm seed laser, one with lasing-off conditions [(a) and blue symbols], the other with lasing-on [(b) and orange symbols]. (a) and (b) LPS measurements after adjusting the timing. (c) Time-resolved energy spread and (d) the same data mapped onto the modulation phase, averaged over about 30 periods. Gray dashed lines in (c) and (d) trace the energy modulation.

the measurement resolution [46]. However, the effect is significantly smaller than the energy spread increase due to lasing. The data indicate that the energy spread increase occurs predominantly between the falling edge and the minima of the energy modulation, which is consistent with the spectral observations presented in End Matter (Appendix C), as well as ML-SASE simulations [47]. Considering that the FEL power is proportional to the square of the energy spread increase [42] and assuming a Gaussian pulse shape, we estimate the FWHM duration of the FEL power profile spikes to be 850 ± 170 as. This is an upper limit, as it is significantly affected by time resolution (estimated around 600 as) and possibly by slippage effects. For comparison, the measured FWHM spectral bandwidth

of 5.9 eV corresponds to a pulse duration of 310 as, assuming transform-limited pulses.

Next we discuss the energy spread based on the phase ϕ_{fit} by averaging over the three nearest slices. The averaged data for ≈ 30 periods at the center of the electron bunch are shown in Fig. 4(d). When lasing is on, the slice energy spread increases more for the slices with phases from 180° to 270° . This is, again, consistent with previous findings. The lasing-on data presented in Fig. 4 are from a single shot. About 50% out of a total of 30 shots were found to show a similar pattern. At this high resolution, the streaking is very sensitive to the electron beam trajectory. Thus, the reason for not observing the behavior in all shots is likely that for some the electron beam and TDS were not aligned.

In summary, we have demonstrated ML-SASE using frequency and time domain probes. This XFEL mode generates trains of periodic, phase-correlated, and subfemtosecond x-ray pulses that exhibit frequency combs. Using a 790-nm seed laser, we have measured a comb frequency jitter of 0.8% (13 meV) and a comb position jitter of 0.03% (0.14 eV). The central photon energy can be controlled with the undulator field, and the period of the train can be tuned with the seed laser wavelength and chicane strength, shown here for 790 and in the End Matter for 263 nm.

The dispersion of the standard chicanes used here may cause overbunching and prevent the scheme from reaching much higher photon energies. This could be overcome by using isochronous chicanes (without dispersion) and by reducing the energy spread of the electron beam. Future efforts at SwissFEL will be dedicated to enhance further the coherence of the output radiation, by using more chicanes and/or by reducing the electron pulse duration, and to perform proof-of-principle phase-sensitive experiments using ML-SASE, which can provide complementary evidence for phase locking.

More broadly, the availability of x-ray pulses with such characteristics is beneficial for a wide range of techniques, including attosecond pump-probe experiments [48,49], frequency comb [50] and time-domain spectroscopies [29,51], and waveform reconstruction by interference [52–55]. The demonstration is a first step toward arbitrary pulse shaping using tailored optical beams for seeding, with the prospect of coherent quantum control and readout of core level excitations. Moreover, we can envision four- or five-dimensional, element-specific imaging techniques which leverage the x-ray frequency comb's prospects to produce attosecond pulses with unparalleled phase stability [56].

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Data availability—The data that support the findings of this article are openly available [57].

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End Matter

Appendix A: MC- and ML-SASE results using a 263.3-nm seed laser—The 263.3-nm seed laser has a FWHM duration of ≈ 195 fs and a pulse energy of ≈ 0.1 mJ. We use fourteen undulator modules to reach close to FEL saturation. More modules are needed for this case, compared to a 790 nm seed laser configuration, due to a reduced optical klystron effect for smaller delays [58]. The achieved pulse energies are

≈ 184 μ J for MC-SASE and ≈ 40 μ J for ML-SASE at 263.3 nm. The spectral results are displayed in Fig. 5. The comparison between MC- and ML-SASE in this case shows similar features to the results with the 790-nm seed laser [Figs. 2(a)–2(d)].

Appendix B: Consecutive single-shot spectra—Figure 6 shows ten consecutive single-shot spectra for MC- (blue) and ML-SASE (orange) with a 790-nm seed laser [the same data as in Figs. 2(a) and 2(b)]. The gray dotted lines in the figure show the background level of the measurement, determined by the averaged intensity of the field-free region. The black lines show the fitted results using Eq. (1). The corresponding fitted contrast values r are shown in the figure, confirming the observation that more than 50% of the ML-SASE spectra show full spectral modulation ($r = 1.0$).

Appendix C: Dependence of the spectral properties on the chicane delay and the seed laser pulse energy—The results of MC- and ML-SASE upon scanning the chicane delay and the seed laser pulse energy are shown in Fig. 7. For the delay scan, the ML-SASE integrated spectral intensity shows a resonant behavior, reaching maximal intensity when the delay approximately

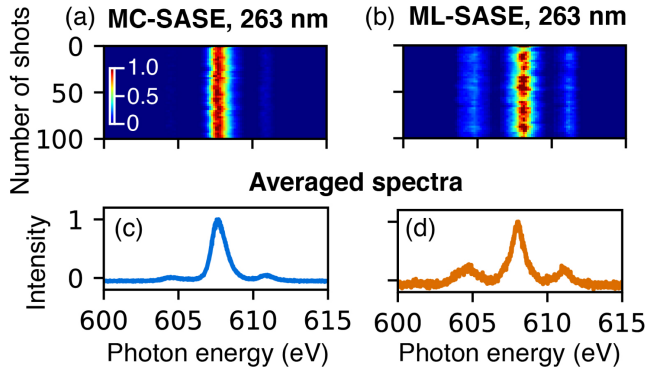


FIG. 5. (a)–(d) Spectral measurements for a 263.3-nm seed laser equivalent to Figs. 2(a)–2(d).

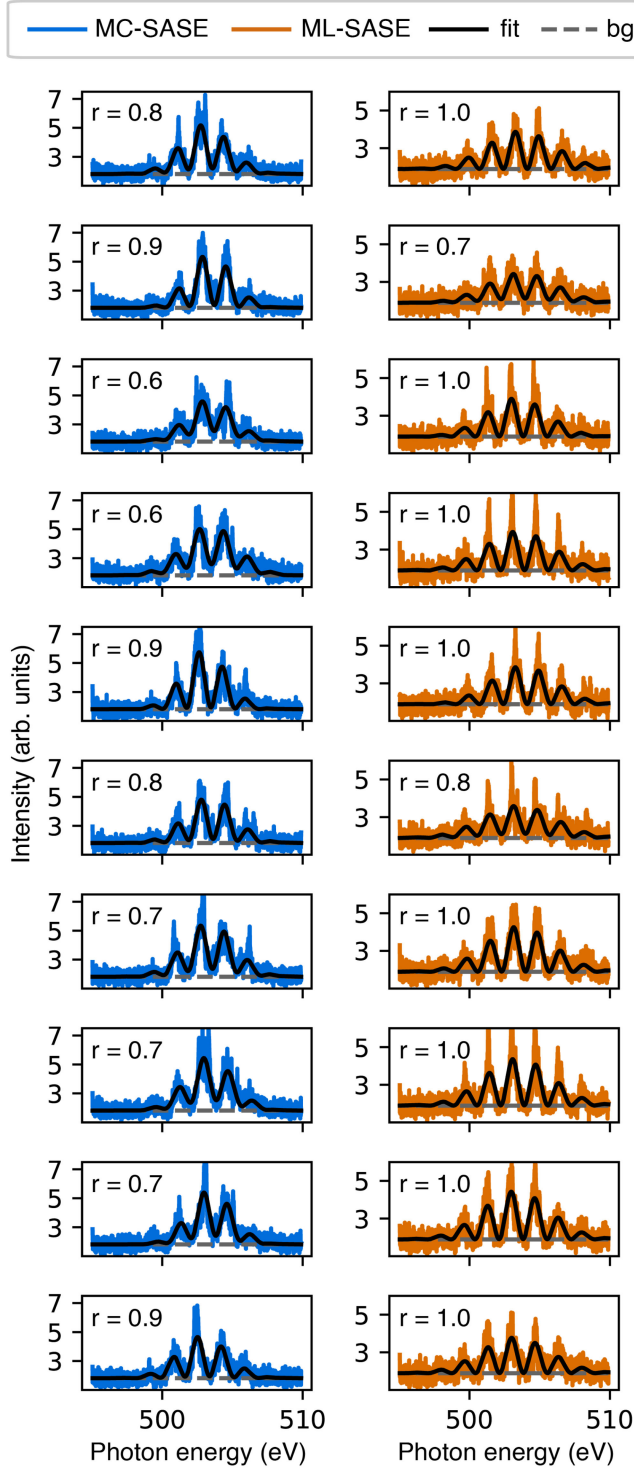


FIG. 6. Ten consecutive single-shot spectra for MC- (blue) and ML-SASE (orange) with a 790-nm seed laser. The black lines show the fit result using Eq. (1). The gray dotted lines show the background level which is identified by the averaged intensity of the field-free region and has been subtracted in the main text. The inserted numbers show the fitted contrast r for the corresponding shots.

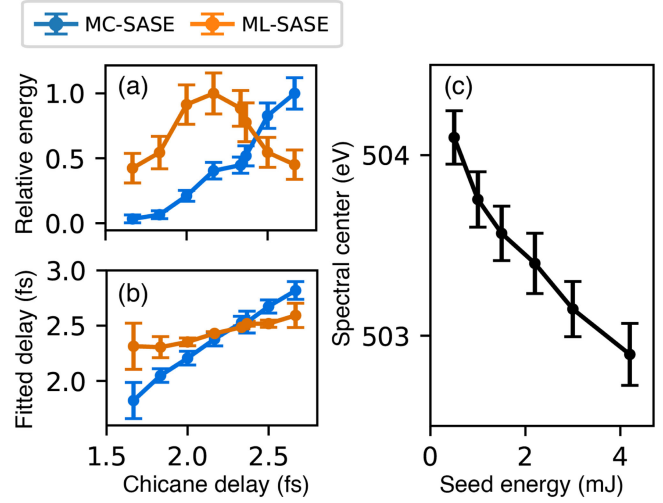


FIG. 7. (a) and (b) show the relative integrated spectral intensity and spectral delay, respectively, as a function of the chicane delay for MC- and ML-SASE. (c) Central photon energy as a function of the seed laser energy.

matches the seed laser wavelength (minus slippage), while for MC-SASE the intensity increases with larger delays due to the optical klystron effect [27,58] (since here the energy spread of the electron beam is rather low, the optimal chicane delay would be larger than the longest measured delay). Moreover, the delays in the spectra [t_0 in Eq. (1)] vary less with ML- than MC-SASE. Additionally, for ML-SASE the shot-to-shot jitter (error) of the fitted delays is reduced when the chicanes match the seed laser wavelength (and vice versa), while for MC-SASE the jitter is found to be rather constant. For the seed laser pulse energy scan, we observe a reduction of the central photon energy of the spectra [obtained from Eq. (1)] upon increasing the laser pulse energy. As the undulator parameters are not changed during the scan, a reduction of the photon energy corresponds to a reduction of the energy of the lasing electrons. Since the laser-induced energy modulation is expected to be proportional to the laser field—namely, a square-root dependence on the seed laser energy—the behavior is consistent with the FEL process occurring for electrons with negative energy modulation [47]. With 4.2 mJ seed laser energy used during the scan [corresponding to an rms energy spread increase of 4 MeV, see Fig 1(c)], the maximal change in electron beam energy is about $\sqrt{2} \times 4 \text{ MeV} / 3.2 \text{ GeV} = 0.18\%$, which corresponds to a 0.36% change in central photon energy. For comparison, the measured change in photon energy is 1.3 eV/504 eV = 0.23%, indicating that the lasing regions are close to the maximal negative modulation.