

Coherent soft x-ray attosecond waveform synthesis through echo-enabled harmonic generation free-electron lasers

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Attosecond pulse trains (APTs) are indispensable for probing electron dynamics at their intrinsic timescales. High-harmonic generation (HHG) has long been a successful and widely used technique in producing extreme ultraviolet APTs. However, in the soft x-ray regime, HHG suffers from low conversion efficiency and lacking flexibility in the waveform and spectrum control. Here in this study, based on the waveform synthesis of the echo-enabled harmonic generation (EEHG) free-electron laser (FEL), we propose a method that can generate soft x-ray APTs with high temporal and spectral tunability. EEHG scheme is well-known in forming phase-locked high-harmonic bunching combs down to the soft x-ray regime. Here, the EEHG scheme is employed to facilitate successive FEL lasing in several different harmonic numbers and then perform the coherent waveform synthesis to generate soft x-ray APTs. Three-dimensional simulation results indicate that reproducible generation of coherent APTs in the soft x-ray regime with peak power of 3.5 GW and micropulse duration of 160 as can be achieved by five-harmonic synthesis. The micropulse duration together with the spectral components of the APTs can be adjusted easily according to the programmable high-harmonic bunching combs in the EEHG scheme. This method has the potential to establish a robust platform for attosecond sciences in the soft x-ray regime, which can enable unprecedented studies of electron dynamics in physical and chemical reactions, biological systems, and quantum materials.

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I. INTRODUCTION

Direct observation and control of electron dynamics at intrinsic timescales drive pioneering research at the intersection of physics, chemistry, and materials science [1,2]. It has provided access to, e.g., photoionization delay, molecular charge migration, electron-electron correlation, electron tunneling, Auger process, and so on [3–5]. Attosecond (1 as = 10^{-18} s) pulse trains (APTs) are ideal tools in the study of these phenomena, which can enable real-time tracking of fundamental quantum processes, providing unparalleled understanding on physical and chemical reactions and material properties [6,7]. High-harmonic generation (HHG) technique has been successful and widely accessible in generating APTs, which

is actually the enabling technique for attosecond sciences [8,9]. HHG now can provide $\sim$ nJ-level pulse energy perharmonic in the extreme ultraviolet (EUV) regime, while in the soft x-ray region, HHG-based APTs face severe problems like low conversion efficiency, high medium absorption, and great phase-matching difficulties [10–12]. These limitations are further compounded by metrological challenges in fully characterizing soft x-ray APTs [13], which is an urgent need for emerging applications from attosecond near-edge x-ray absorption fine structure spectroscopy to single-shot coherent diffraction imaging [14,15]. Extending APTs into the soft x-ray region is also essential for element-specific probing of core-level transitions, high spatiotemporal resolution imaging, and reduced radiation damage in biological samples [16,17]. These limitations fundamentally constrain HHG-based APTs to the EUV regime, leaving a critical gap for high-flux, flexible soft x-ray attosecond sources needed for element-specific spectroscopy and reduced-damage biological imaging.

Besides HHG sources, free-electron lasers (FELs) are rapidly advancing toward the generation of APTs [18,19], isolated attosecond pulses (IAPs) [20–24], and attosecond sciences [25–27]. FELs employ high energy relativistic electron beams in undulator magnets to generate ultrahigh

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brightness, ultrashort pulse duration, and wavelength-tunable radiation pulses from EUV to x-ray regime. Most FEL facilities are working on the basis of self-amplified spontaneous emission (SASE) scheme [28,29], in which the FEL longitudinal coherence is fundamentally constrained by the electron beam shot noise and the radiation slippage effects [30]. Seeded FEL schemes such as high gain harmonic generation (HGHG) [31,32] and echo-enabled harmonic generation (EEHG) [33,34] overcome the limitation and improve the longitudinal coherence by imprinting fully coherent optical laser into the electron beam. In EEHG, two laser modulators and dispersive sections are employed to form a fine-structured longitudinal phase space distribution in the electron beam, yielding fully coherent high-harmonic bunching combs with well-defined relative phases. Due to the finer bunching structures, EEHG has higher frequency-up-conversion efficiency than HGHG, which can easily reach the soft x-ray region now.

Several FEL-based methods for generating high-power APTs at short wavelength have been proposed [18,19,35–39]. Among them, LCLS has successfully demonstrated the incoherent attosecond x-ray pulses using SASE-based approaches [19], and free electron laser radiation for multidisciplinary investigations (FERMI) has demonstrated the coherent APTs' generation in the EUV region [18,39], by employing the waveform synthesis (WS) technique [40] into the HGHG-FEL scheme. In general, the WS technique is widely used in the optical community for attosecond scientific researches [41,42]. Advanced study results reveal that, combining the WS technique with high gain FEL principles, APTs' generation with short pulse duration down to even zeptosecond, high photon energy beyond 10 keV and lots of scientific applications can be made possible [38,39,43]. The main principle of the WS technique is the coherent superimposition of multiple phase-locked optical fields. Hence, it can be easily implemented in seeded FEL schemes like the HGHG and the EEHG.

Despite these breakthroughs, extending attosecond WS to the soft x-ray regime with deterministic multiline spectral composition remains challenging: HHG suffers from limited flux and restricted control in soft x-rays, SASE lacks intrinsic shot-to-shot phase reproducibility, and HGHG faces practical limitations in reaching very high harmonics with a freely tailorable harmonic comb. Hence in this study, we propose a method to generate APTs in the soft x-ray regime with high spectral and temporal tunability by leveraging the advanced EEHG scheme and combining it with the WS technique. The unique advantage of the EEHG scheme means we can easily achieve short wavelength FEL lasing down to the soft x-ray region and even cover the “water-window” regime [44,45]. And the high-harmonic bunching spectrum in EEHG can be controlled intentionally and tuned easily, which helps us gain another advantage of flexible waveform and spectrum control. By strategically controlling and optimizing of parameters like modulation strengths, chicane dispersions, and phase shifters, three-dimensional simulation results prove that, through our method, phase-locked high-harmonic FEL lasing pulses in several different harmonic numbers can be obtained, coherent waveform synthesis can be performed, and soft x-ray APTs' generation can be eventually achieved. For five-harmonic synthesis, APTs in the soft x-ray region with peak power of 3.5 GW and micropulse duration of 160 as can

be expected. The flexible waveform and spectrum control is also demonstrated in this study. The proposed method shows great potential in the generation and application of fully coherent soft x-ray APTs. In principle, the underlying mechanism can be extended from the ultraviolet to the “water window.”

II. PRINCIPLE

The schematic layout of the proposed method is illustrated in Fig. 1, comprising three key functional sections: (1) two-stage modulation-dispersion sections (M1, C1, M2, C2) and two external seed lasers (Seed1, Seed2) to implement the EEHG scheme, (2) multisegment radiators (R1, R2, R3, ...) for FEL lasing in distinct high harmonics, and (3) phase shifters (PS1, PS2, ...) positioned between successive radiator segments to enable precise relative phase control for the WS. In operation, a high-quality electron beam from the linac first traverses the EEHG scheme to acquire the high-harmonic microbunching imprinted by the seed lasers. The prebunched electron beam then propagates through the radiator segments, generating coherent FEL radiation pulses at multiple harmonics with fixed phase relationships. This intrinsic phase coherence renders WS via coherent superposition feasible. Crucially, the phase shifters between radiator stages provide fine-tuning of relative phases required for stable and programmable WS, as demonstrated in prior work [46]. The underlying principles and implementation details are presented in the following subsections.

A. High-harmonic bunching generation and control in the EEHG scheme

The EEHG scheme employs sequential interactions between the electron beam and external seed lasers (Seed1, Seed2) in modulators M1 and M2 to imprint energy modulations into the electron beam. The first magnetic chicane (C1), characterized by a strong longitudinal dispersion, stretches the energy modulation in M1 into finely structured energy bands in the electron beam's longitudinal phase space. A second energy modulation is then applied in M2, superimposed on this prestructured energy distribution. Subsequently, the weaker dispersion of chicane C2 converts the combined energy modulation into a high-fidelity density modulation. Owing to the fine granularity of the initial energy bands, this process generates pronounced microbunching at very high harmonics of the seed laser frequency.

A distinctive advantage of the EEHG scheme lies in its ability to generate very high-harmonic microbunching using only modest energy modulation amplitudes. The resulting harmonic bunching spectrum follows the theoretical framework established by Stupakov [33], Xiang and Stupakov [34], and Kim *et al.* [47], in which the complex bunching factor at harmonic order h is given by

$$b_h = e^{im\phi} \exp \left[-\frac{(hB_2 - B_1)^2}{2} \right] J_m(-hA_2B_2) \times J_{-1}[-A_1(-B_1 + hB_2)]. \quad (1)$$

Here, ϕ represents the relative phase between Seed1 and Seed2, J_m denotes the m th order Bessel function of the first kind, and the harmonic number is given by $h = m\kappa - 1$ with

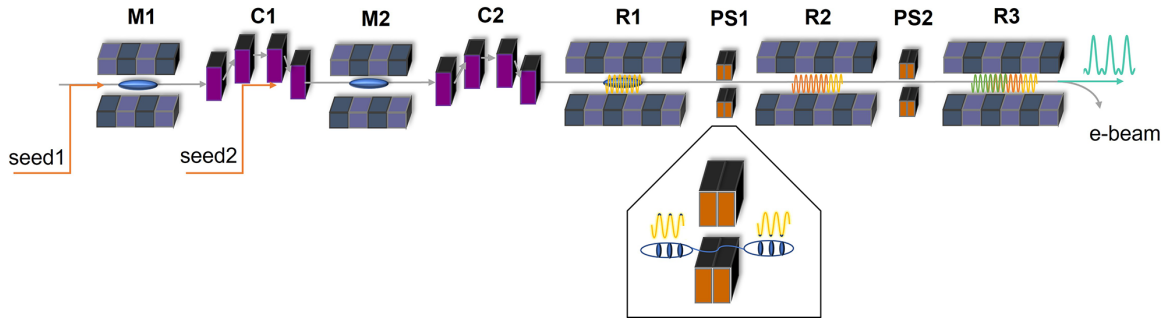


FIG. 1. Schematic layout of the proposed method, mainly composing an EEHG-FEL setup and a waveform synthesis configuration. M1 and M2 denote the first and second modulators, C1 and C2 the corresponding dispersive chicanes, R1–R3 the radiator segments tuned to different harmonic numbers, and PS the phase shifters placed between successive radiators. Solid black lines represent the electron beam trajectory, while colored wavy lines indicate the generated FEL radiation at different harmonics. The arrows highlight the sequence of modulation, dispersion, harmonic amplification, and coherent waveform synthesis.

$\kappa = \lambda_1/\lambda_2$. Here, λ_1 and λ_2 are the wavelengths of Seed1 and Seed2, respectively, and $k_1 = 2\pi/\lambda_1$ and $k_2 = 2\pi/\lambda_2$ are the corresponding wave numbers. The dimensionless modulation and dispersion parameters are defined as

$$A_1 = \frac{\Delta E_1}{\sigma_E}, \quad A_2 = \frac{\Delta E_2}{\sigma_E},$$

$$B_1 = \frac{R_{56}^{(1)} k_1 \sigma_E}{E_0}, \quad B_2 = \frac{R_{56}^{(2)} k_2 \sigma_E}{E_0},$$

where ΔE_1 and ΔE_2 are the energy modulation amplitudes imparted in modulators M1 and M2, $R_{56}^{(1)}$ and $R_{56}^{(2)}$ are the longitudinal dispersion strengths of chicanes C1 and C2, σ_E is the initial rms energy spread of the electron beam, and E_0 is the nominal beam energy. It follows from Eq. (1) that the magnitude of the EEHG bunching factor, $|b_h|$, is primarily governed by the four dimensionless parameters (A_1, A_2, B_1, B_2), enabling precise control over the harmonic content through tailored modulation and dispersion settings.

To illustrate the characteristic effects of the EEHG scheme, we present the typical longitudinal phase space, corresponding current profile, and harmonic bunching spectrum in Fig. 2, simulated with modulation and dispersion parameters $A_1 = 6.5$, $A_2 = 5.6$, $B_1 = 6.1$, and $B_2 = 0.20$. Figure 2(a) shows a longitudinal slice of the electron beam spanning one

seed laser wavelength, and the corresponding current profile is shown in Fig. 2(b). The fine-scale structure evident in this slice confirms the generation of very high harmonic content through the EEHG process. The harmonic bunching spectrum is shown in Fig. 2(c), exhibiting a peak of nearly 11% at the 32nd harmonic of the seed laser—achieved using only moderate modulation amplitudes ($A_1, A_2 \sim 5$ –6). Notably, the EEHG bunching spectrum exhibits a distinct, nonmonotonic pattern, in stark contrast to the monotonic (approximately exponential) decay observed in the HGHG scheme [48]. This structured spectral envelope provides an additional degree of freedom for tailoring harmonic content.

In general, the EEHG scheme enables selective enhancement of a target harmonic by optimizing the modulation and dispersion parameters. Crucially, these parameters govern distinct aspects of the harmonic bunching spectrum: The first-stage energy modulation amplitude A_1 primarily shapes the overall envelope and peak intensity distribution of the harmonic bunching, while the first-stage dispersion parameter B_1 determines the harmonic order at which maximum bunching occurs. Figure 3 summarizes the evolution of the harmonic bunching spectrum across harmonic orders under independent scans of A_1 and B_1 , with other parameters remain the same as in Fig. 2. As shown in Fig. 3(a), decreasing the first-stage modulation amplitude A_1 broadens the harmonic comb and

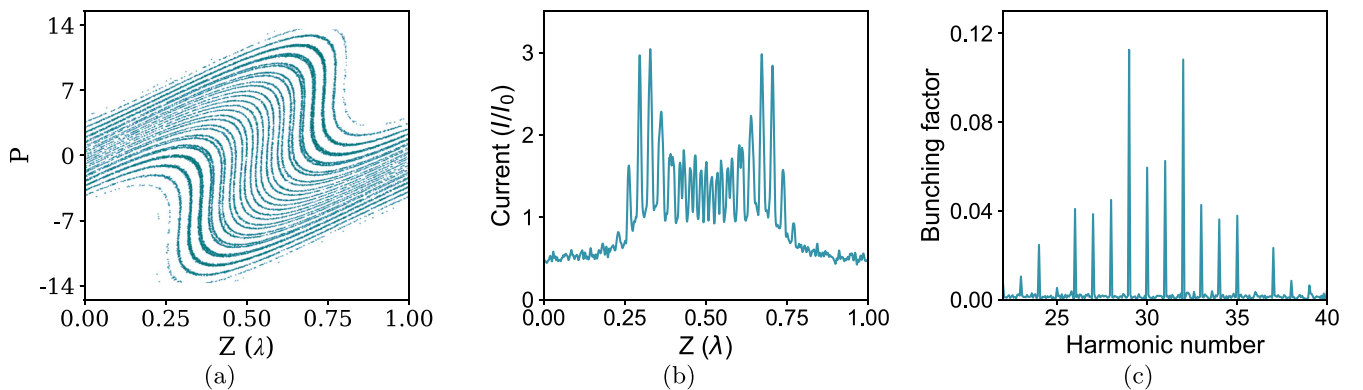


FIG. 2. Typical longitudinal phase space [panel (a)], current profile [panel (b)], and high-harmonic bunching factor $|b_n|$ [panel (c)] for the EEHG scheme with $A_1 = 6.5$, $A_2 = 5.6$, $B_1 = 6.1$, and $B_2 = 0.20$. In panel (a), Z denotes the longitudinal coordinate normalized to the seed laser wavelength λ_{seed} , and P represents the normalized relative energy deviation defined as $P = (E - E_0)/\sigma_E$.

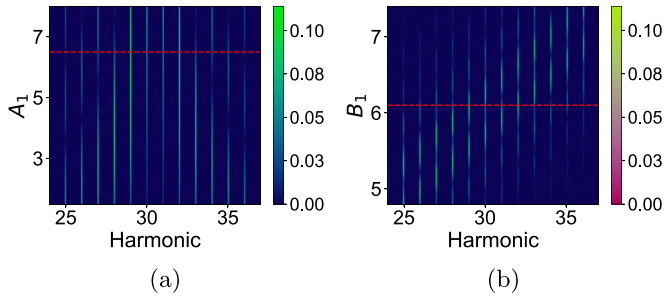


FIG. 3. Evolution of the harmonic bunching spectrum with the variation of EEHG parameter. (a) Dependence on the first-stage modulation amplitude A_1 , with $A_2 = 5.6$, $B_1 = 6.1$, and $B_2 = 0.20$ fixed. (b) Dependence on the first-stage dispersion parameter B_1 , with $A_1 = 6.5$, $A_2 = 5.6$, and $B_2 = 0.20$ fixed. The red dashed lines correspond to the parameter sets in Fig. 2.

enhances bunching in the sideband harmonics, effectively increasing the spectral bandwidth of the microbunching. In contrast, increasing A_1 concentrates the bunching strength into fewer harmonics, yielding a narrower but more intense spectral envelope. Varying B_1 shifts the target harmonic with the peak bunching factor without significantly altering the overall amplitude, as illustrated in Fig. 3(b), which means this can enable precise tuning of the optimal harmonic number easily. These results demonstrate that the EEHG process provides two orthogonal degrees of freedom for harmonic control: A_1 for spectral shaping and intensity scaling, and B_1 for selecting the target harmonic. This dual-parameter programmability establishes a robust foundation for flexible spectral and temporal waveform engineering in the subsequent WS processes.

B. FEL lasing and harmonic bunching evolution in multistage radiators

Following the generation of high-harmonic bunching combs in the EEHG stage, the electron beam propagates through a series of multistage radiators to undergo coherent FEL amplification at different harmonic numbers, which is prerequisite for the subsequent WS processes. Precise control over the amplification dynamics and evolution of the bunching combs in multistage radiators is therefore essential. Figure 4 illustrates a representative example of harmonic bunching evolution during the FEL lasing process in a radiator tuned

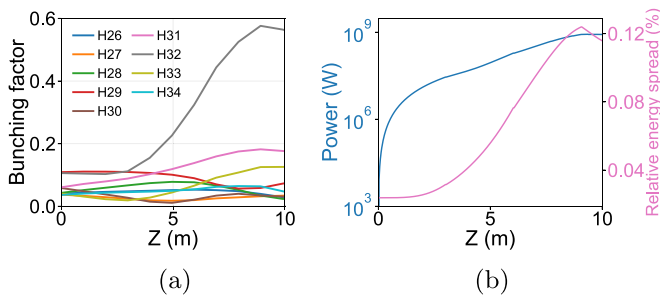


FIG. 4. Evolution of (a) all harmonics bunching factor and (b) the power and energy spread during the FEL lasing process. The radiator is tuned to resonate at the 32nd harmonic.

to a single resonant wavelength, initialized with the same bunching spectrum as in Fig. 2(c). The radiator is tuned to resonance at the 32nd harmonic (H_{32}), ensuring optimal overlap between the electron beam's microbunching structure and the radiation field. As the beam traverses the radiator, the seeded FEL process initiates coherent energy transfer from the electrons to the radiation field at this harmonic. This interaction not only amplifies the radiation power but also reinforces the electron density modulation through the FEL ponderomotive potential—a positive feedback mechanism known as microbunching enhancement. Consequently, the bunching factor of the 32nd harmonic increases from an initial value of approximately 11% [inherited from the EEHG stage, cf. Fig. 2(c)] to over 58% at saturation, while the FEL power grows to the gigawatt level and the relative energy spread rises from 0.02% to 0.12%. This coherent exponential gain process significantly enables high-power FEL lasing and strengthens the electron beam microbunching.

In addition, Fig. 4(a) reveals that the initially strong bunching factor of the 30th harmonic (H_{30}) gradually decreases and the bunching factor of 31st harmonic (H_{31}) gradually increasing to nearly 15% at $z = 8$ m. The reason is that, while the initial harmonic bunching spectrum is tailored by the modulation and dispersion parameters (A_1 , A_2 , B_1 , B_2), the longitudinal dispersion R_{56} introduced in the radiator further modulates the bunching evolution. Specifically, this radiator dispersion also amplifies lower-order harmonics near the resonant frequency (here, H_{32}) and facilitates smooth gain transfer among neighboring harmonics. This dispersion-engineered bunching dynamics enables the sustained amplification of multiple high harmonics along the radiator line. Consequently, in a multistage radiator configuration, successive segments can be tuned to lase at different harmonic numbers—such as H_{32} , H_{31} , and H_{30} —producing a set of phase-locked radiation pulses. These pulses will serve as the essential building blocks for WS and the subsequent generation of APTs.

It is important to note that the significant growth in FEL radiation power will inevitably increase the electron beam's energy spread. This enlarged energy spread will hence degrade the microbunching fidelity and suppress further FEL amplification in downstream radiator sections. Consequently, for each harmonic lasing stage, both the saturation power and the radiator length must be carefully optimized to balance high output power against beam quality preservation. Furthermore, the longitudinal dispersion R_{56} that can be practically introduced in the radiator segments is inherently limited by the overall saturation length, and excessive dispersion would distort the electron beam's dynamics. This constraint restricts the spectral window over which neighboring harmonics can be simultaneously amplified through dispersion engineering, thereby defining an upper bound on the harmonic comb bandwidth achievable in a single radiator stage. Based on the principles and analysis outlined above, a five-segment radiator system—tuned sequentially to H_{32} through H_{28} with tailored segment lengths—can be implemented. The evolution of harmonic bunching across these radiator sections is shown in Fig. 5. As previously discussed, the bunching factor of lower-order harmonics near the resonant harmonic is enhanced during FEL amplification, enabling their subsequent lasing in downstream radiator stages. For instance, radiator section R1

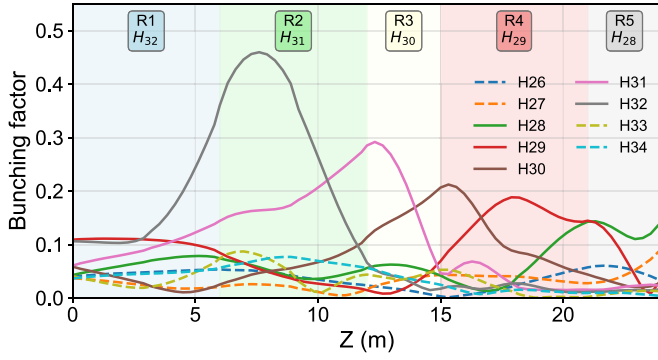


FIG. 5. Dispersion-enhanced high-harmonic bunching evolution along the five-segment radiator system for five-harmonic synthesis. R_1 (0–6 m, blue) is tuned to resonate at the 32nd harmonic (H_{32}); R_2 (6–12 m, green) is tuned to resonate at the 31st harmonic (H_{31}); R_3 (12–15 m, yellow) is tuned to resonate at the 30th harmonic (H_{30}); R_4 (15–21 m, red) is tuned to resonate at the 29th harmonic (H_{29}); R_5 (21–24 m, gray) is tuned to resonate at the 28th harmonic (H_{28}). The design of longitudinal dispersion (R_{56}) also enables selective enhancement of lower-order harmonics in each radiator segment, thereby achieving multiharmonic cascaded amplification. Note: For clarity, the figure shows only the evolution of the relevant harmonics (H_{28} – H_{32}).

is tuned to (H_{32}). During FEL amplification in R_1 , not only does the bunching factor of H_{32} grow significantly, but that of the adjacent H_{31} also increases substantially. This enhanced H_{31} microbunching then serves as the seed for FEL lasing in the next radiator section, R_2 . This cascaded amplification process can be repeated across multiple stages, provided that the bunching structure remains intact and the electron beam retains sufficient coherence. However, the process eventually terminates when either the EEHG-induced microbunching structure is disrupted, causing the bunching factor of the next target harmonic to become negligible, or the energy spread of the electron beam is significantly increased by prior FEL interactions, thereby suppressing further amplification. These results demonstrate that judicious dispersion management, combined with multistage FEL amplification, enables flexible, configuration-dependent shaping of the harmonic combs. Specifically, the synergistic integration of the EEHG parameters (A_1, A_2, B_1, B_2), longitudinal dispersion R_{56} , and tailored multisegment radiators collectively facilitate dynamic control over high-harmonic bunching evolution. This capability constitutes the core methodological innovation underlying the programmable soft x-ray WS presented in the following sections.

C. Coherent multicolor WS for APTs' generation

With a programmable harmonic comb and multiharmonic FEL lasing established, we can then perform the WS technique to generate APTs through the coherent superposition of the radiation fields. The temporal profile of the synthesized pulses is governed by four controllable parameters: the number of harmonics (N), their spectral amplitudes (M_k), angular frequencies (ω_k), and phases (ϕ_k). Precise phase matching among these harmonics is achieved via phase shifters

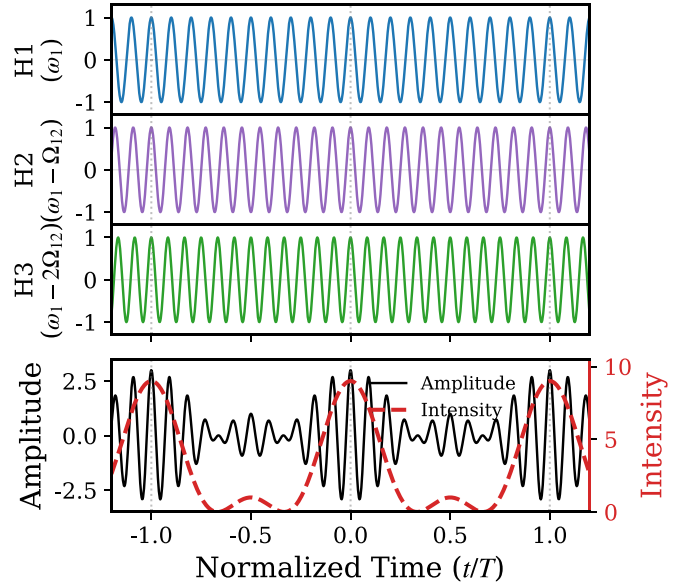


FIG. 6. Mechanism of coherent harmonic synthesis. The top three panels display the temporal oscillation of three individual harmonic components with frequencies ω_0 (blue), $\omega_0 - \Omega_{12}$ (purple), and $\omega_0 - 2\Omega_{12}$ (green). Phase locking ensures that all harmonic waves constructively align at $t = 0$, as indicated by the vertical dotted line. The bottom panel shows the synthesized waveform resulting from the superposition of these harmonics. The solid black curve represents the total electric field amplitude (left axis). The dashed red curve depicts the intensity (right axis), illustrating the formation of a high-intensity pulse due to the constructive interference of the spectral components. The time axis is normalized to the beat period $T = 2\pi/\Omega_{12}$.

integrated between successive radiator segments. This is essential for stable WS. The phase matching is achieved by tuning the deflection parameter K of the phase shifters. The resulting phase shift $\Delta\phi$ is given by

$$\Delta\phi = \frac{2\pi}{\lambda} \frac{L_{ps}}{2\gamma^2} \left(1 + \frac{K^2}{2}\right), \quad (2)$$

where λ is the resonant wavelength of the target harmonic, L_{ps} denotes the effective length of the phase shifter, and γ is the relativistic Lorentz factor.

To elucidate the underlying generation mechanism, we define the instantaneous radiation intensity as $I_{inst}(t)$. According to Poynting's theorem, this intensity can be expressed as

$$I_{inst}(t) = \epsilon_0 c E_{total}^2(t), \quad (3)$$

where the total electric field comprises N harmonic components:

$$E_{total}(t) = \sum_{k=1}^N M_k e^{i(\omega_k t + \phi_k)}. \quad (4)$$

To aid clarity, Fig. 6 schematically illustrates the temporal waveform evolution resulting from the coherent superposition of harmonic components discussed below. For closely spaced harmonics ($\Delta\omega \ll \bar{\omega}$), the rapidly oscillating carrier terms average out over observation timescales $\tau \gg 2\pi/\bar{\omega}$, yielding

the slowly varying envelope intensity:

$$I_{\text{env}}(t) = \frac{\epsilon_0 c}{2} \left[\sum_{k=1}^N M_k^2 + 2 \sum_{i < j} M_i M_j \cos(\Omega_{ij} t + \Phi_{ij}) \right], \quad (5)$$

where $\Omega_{ij} = \omega_i - \omega_j$ and $\Phi_{ij} = \phi_i - \phi_j$ denote the frequency and phase differences between harmonics i and j , respectively.

The maximum intensity is attained when all interference terms constructively interfere, i.e., when $\cos(\Omega_{ij} t + \Phi_{ij}) = 1$, for all $i < j$:

$$I_{\text{max}} = \frac{\epsilon_0 c}{2} \left(\sum_{k=1}^N M_k \right)^2. \quad (6)$$

This condition requires the relative phases to satisfy a linear relationship with the frequency differences, which means equivalently a constant group delay (i.e., zero group-delay dispersion) across the harmonic comb:

$$\frac{\Phi_{ij}}{\Omega_{ij}} = \tau_0 \quad (\tau_0 \in \mathbb{R}), \quad (7)$$

which will ensure temporal synchronization of all harmonic components. Based on Eq. (7), random phase jitter in the phase shifters directly translates to deviations in the relative phases Φ_{ij} . A quantitative analysis shows that maintaining the phase fluctuations below approximately 0.1 rad is sufficient to preserve the coherent superposition required by the proposed waveform synthesis scheme [43,49]. The permanent-magnet phase shifters of Shanghai soft x-ray free-electron laser (SXFEL) shows an operating accuracy of below 2 mrad, which can satisfy the requirements of Eq. (7). Under this phase-matching condition, the synthesized waveform becomes strictly periodic, with pulse period determined by the greatest common divisor of all pairwise frequency differences:

$$T = \frac{2\pi}{\text{gcd}(\{\Omega_{ij}\})}. \quad (8)$$

To quantify the degree of coherent enhancement achieved through WS, we define the peak intensity enhancement factor as

$$\alpha = \frac{I_{\text{max}}}{\sum_{k=1}^N I_k} = \frac{(\sum_{k=1}^N M_k)^2}{\sum_{k=1}^N M_k^2}. \quad (9)$$

Under ideal phase matching, the minimum intensity can vanish if the largest amplitude does not dominate the sum of the others, i.e., if $M_{\text{max}} \leq \sum_{k \neq \text{max}} M_k$. Otherwise, the intensity is bounded below by

$$I_{\text{min}} \geq \frac{\epsilon_0 c}{2} \left(2M_{\text{max}} - \sum_{k=1}^N M_k \right)^2. \quad (10)$$

For dual-harmonic synthesis ($N = 2$), the relative phase $\Phi_{12} = \phi_1 - \phi_2$ can be freely adjusted, and the optimal signal-to-noise ratio (SNR)—defined as the peak-to-sidelobe intensity ratio—is achieved when $M_1 = M_2$, yielding a vanishing minimum intensity and maximal $\alpha = 2$. In contrast,

TABLE I. Simulation parameters based on the SXFEL facility.

Parameter	Value	Unit
Electron beam		
Energy	1.3	GeV
rms energy spread	60	keV
Normalized emittance	1	mm mrad
Peak current	800	A
Seed lasers		
Wavelength	266	nm
Pulse duration (Seed1)	100	fs
Pulse duration (Seed2)	100	fs
Undulator system		
Modulator period (M1, M2)	8	cm
Modulator length (M1, M2)	1.28	m
Radiator period	3	cm
Radiator segment length	3	m
K (R1)	1.61	—
K (R2)	1.64	—
K (R3)	1.68	—
K (R4)	1.72	—
K (R5)	1.76	—

multiharmonic synthesis ($N \geq 3$) requires strict adherence to the linear phase-matching condition in Eq. (7). While the peak intensity enhancement factor α is maximized for equal amplitudes ($M_k = M$), the temporal contrast (SNR) can be further optimized through tailored amplitude shaping. Moreover, equally spaced harmonics are essential to ensure a periodic pulse train with a well-defined repetition rate and high temporal contrast.

III. SIMULATION RESULTS

A. Soft x-ray APTs' generation

To evaluate the proposed method, we performed three-dimensional numerical simulations based on realistic machine parameters from the SXFEL facility [50]. The FEL amplification process was modeled using the time-dependent FEL code GENESIS 1.3 [51]. The electron beam parameters were set to representative operational values: a beam energy of 1.3 GeV, an rms energy spread of 60 keV, a normalized emittance of 1 mm mrad, and a peak current of 800 A. Two external seed lasers at 266 nm were employed, Seed1 and Seed2, both with a pulse duration of 100 fs and derived from the same laser source. The undulator system was optimized for soft x-ray harmonic generation, featuring a period of 3 cm and alternating radiator segments of 3 m length. A complete list of simulation parameters is provided in Table I.

We simulated a five-harmonic WS scheme spanning consecutive harmonics from the H_{32} to the H_{28} . The EEHG stage was configured with the same parameters as in Fig. 2(c), yielding a broad and structured bunching comb with significant amplitudes across the target harmonic range. This initial harmonic bunching spectrum serves as the seed for staged amplification in a five-segment radiator system, whose layout is illustrated in Fig. 5. As the prebunched electron beam propagates through successive radiator sections—each

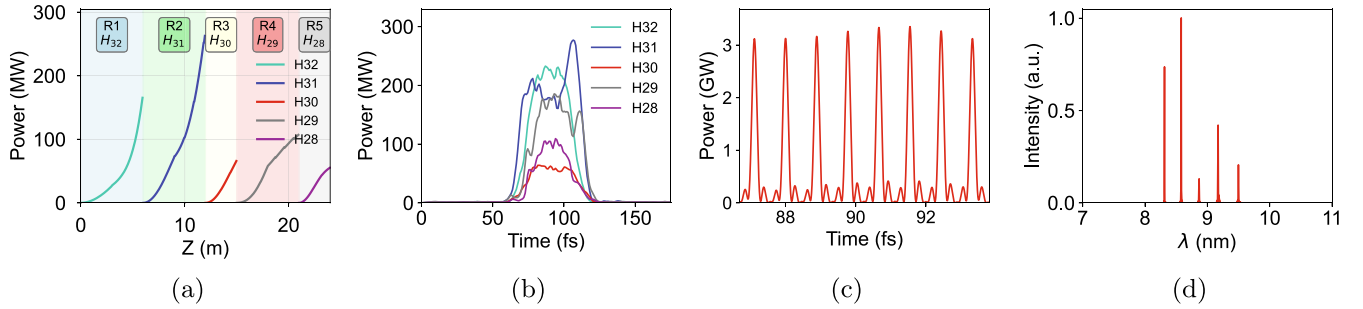


FIG. 7. Five-harmonic WS results for consecutive harmonics from H_{32} to H_{28} : (a) radiation power evolution [R_1 (0–6 m, blue) is tuned to resonate at the 32nd harmonic (H_{32}); R_2 (6–12 m, green) is tuned to resonate at the 31st harmonic (H_{31}); R_3 (12–15 m, yellow) is tuned to resonate at the 30th harmonic (H_{30}); R_4 (15–21 m, red) is tuned to resonate at the 29th harmonic (H_{29}); R_5 (21–24 m, gray) is tuned to resonate at the 28th harmonic (H_{28})] and (b) radiation power profile at radiation exit of individual harmonic number, (c) temporal profile, and (d) phase-locked harmonic components of the synthesized APTs.

tuned to resonate at a specific harmonic (R_1 : H_{32} , R_2 : H_{31} , R_3 : H_{30} , R_4 : H_{29} , R_5 : H_{28})—the interplay between FEL gain and radiator-induced longitudinal dispersion (R_{56}) enables efficient energy extraction into multiple harmonic radiation fields, which means we can get FEL lasing pulses in several different harmonics. Figure 7(b) shows the results of the radiation power profiles at the exit of each radiator section, confirming that all five harmonics FEL radiation pulses reach tens to hundreds of megawatts peak power output. This successful multiharmonic FEL lasing is a direct manifestation of the dispersion-managed bunching evolution described in Sec. II B, where R_{56} in each radiator selectively enhances lower-order harmonics near resonance, thereby facilitating smooth gain handover between successive stages.

The phase coherence imprinted during the EEHG process is preserved throughout the amplification chain, as evidenced by the narrow spectral linewidths of the harmonic fields in Fig. 7(d). This coherence will enable high-fidelity coherent superposition via the WS technique, with all the spectral lines are within the soft x-ray regime. The four phase shifters between the five radiator segments were adjusted to satisfy the linear phase-matching condition in Eq. (7) with $\Delta\Phi_{ij} = \phi_{12} = \phi_{23} = \phi_{34} = \phi_{45} = 0$ (the parameter will become the default in simulations unless stated otherwise), yielding the synthesized APT shown in Fig. 7(c), which exhibits a compressed micropulse duration of 160 as, a repetition rate of 2.25×10^{15} Hz, and a peak power exceeding 3.5 GW. The temporal contrast being nearly 10, defined as the ratio of the main pulse peak intensity to that of the nearest sidelobe, confirms that the synthesized waveform is not only intense but also highly structured, which will fulfill the stringent requirements for phase-sensitive attosecond spectroscopy [9]. The results demonstrate that by synergistically combining programmable EEHG bunching, dispersion-engineered radiator staging, and precise phase control, our method can achieve simultaneous manipulation and FEL lasing of several different harmonics, which is a critical step toward fully customizable soft x-ray APTs. In practical seeded FEL configurations, this consideration typically restricts the number of simultaneously usable harmonics to fewer than about seven. The five-harmonic synthesis demonstrated in this work should therefore be regarded as a representative configuration rather than a unique or generally optimal choice.

B. Waveform and spectrum control

The above results demonstrate successful waveform synthesis and soft x-ray APTs' generation from a consecutive five-harmonic combs. To further highlight the flexibility of waveform and spectrum control in our method, we consider a nonconsecutive harmonic comb by reoptimizing the EEHG parameters to enhance bunching at selected target harmonics. Specifically, we demonstrate triple-harmonic WS by coherently combining the H_{32} , H_{30} , and H_{28} to further validate the waveform tunability in our method. The modulation amplitudes were set to $A_1 = 6.0$ and $A_2 = 4.0$, with normalized dispersion strengths $B_1 = 8.6$ and $B_2 = 0.28$, respectively. This configuration yields pronounced initial microbunching at H_{32} and H_{30} , with bunching factors exceeding 0.11, as shown in Fig. 8(a).

In the first radiator segment (a single-undulator, 3 m), the 32nd harmonic reached a peak power of 35 MW. The 30th harmonic reached a peak power of 85 MW in the second radiator segment (a dual-undulator, 6 m). The third radiator segment also employed a 6 m dual-undulator configuration to enhance radiation efficiency, achieving a peak power of 65 MW at the 28th harmonic. The two phase shifters between the three radiator segments were adjusted to satisfy the linear phase-matching condition in Eq. (7), yielding a synthesized APT with a peak power of 525 MW, a micropulse duration of 190 as, and a temporal contrast of 25. The corresponding temporal profile and phase-locked spectrum are shown

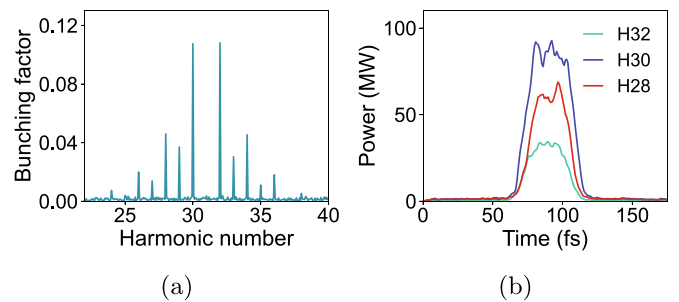


FIG. 8. High-harmonic bunching factor (a) and radiation power of individual harmonics (b) for nonconsecutive harmonic synthesis with H_{32} , H_{30} , and H_{28} . The EEHG parameters are tuned to be $A_1 = 6.0$, $A_2 = 4.0$, $B_1 = 8.6$, and $B_2 = 0.28$ now.

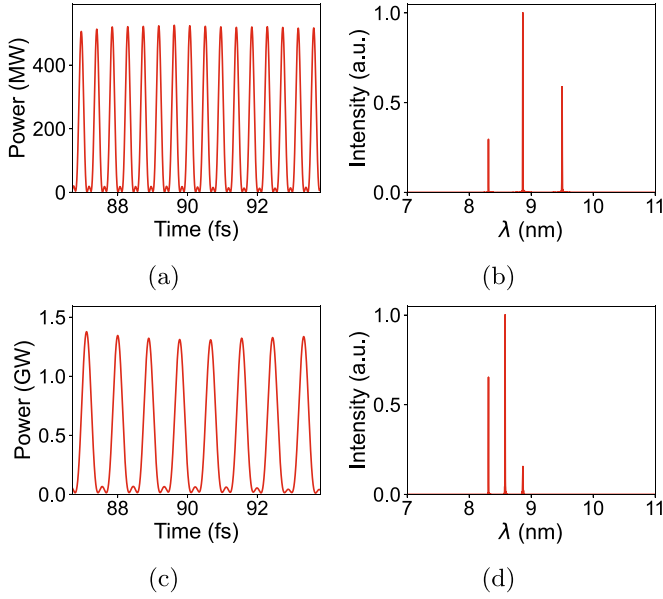


FIG. 9. Power and spectrum profile of the synthesized APTs for triple-harmonic synthesis with nonconsecutive H_{32} , H_{30} , H_{28} [panels (a) and (b)] and consecutive H_{32} , H_{31} , H_{30} [panels (c) and (d)] harmonic numbers.

in Figs. 9(a) and 9(b), respectively. For comparison purpose, we also illustrate the synthesized results of three consecutive harmonics (H_{32} – H_{30}) extracted from the first three radiator stages in Fig. 7. The temporal and spectral results are shown in Figs. 9(c) and 9(d), respectively. One can find out that the micropulse duration here is clearly increased, which is about 335 as comparing to the 190 as in Fig. 9(a). The results presented here mean that we can adjust the waveform of the APTs by synthesis of nonconsecutive harmonic combs in our method.

To further validate the spectral tunability, we adjusted the EEHG dispersion parameter B_1 from 8.6 to 8.9 while retaining the nonconsecutive harmonic layout. We know from Fig. 3(b) that variation of B_1 will shift the high-harmonic bunching spectrum in the EEHG scheme. The modified initial bunching spectrum now is shown in Fig. 10(a), in which we can see the optimal bunching harmonics are H_{33} and H_{31} , which is shifted one-harmonic order larger from H_{32} and H_{30} in Fig. 8(a). Keeping the radiator architecture unchanged, we then retuned the resonance conditions of the three radiator segments to H_{33} , H_{31} , and H_{29} , respectively. The power distribution of high harmonics FEL lasing at the exit of each radiator segment is presented in Fig. 10(b). A comparison with the case in Fig. 8(b) reveals that the output powers of all three harmonics remain the same levels, confirming successful amplification across the shifted harmonic combs. After adjusting the phase shifters here, we can also achieve coherent WS of the H_{33} , H_{31} , and H_{29} harmonics. The resulting temporal and spectral characteristics are shown in Figs. 10(c) and 10(d). Comparing them with the baseline synthesis results in Figs. 9(a) and 9(b), one can find out that the synthesized waveform retains a nearly identical pulse structure and micropulse duration, while the entire spectrum is shifted to shorter wavelengths accordingly. These results prove that the proposed scheme offers excellent

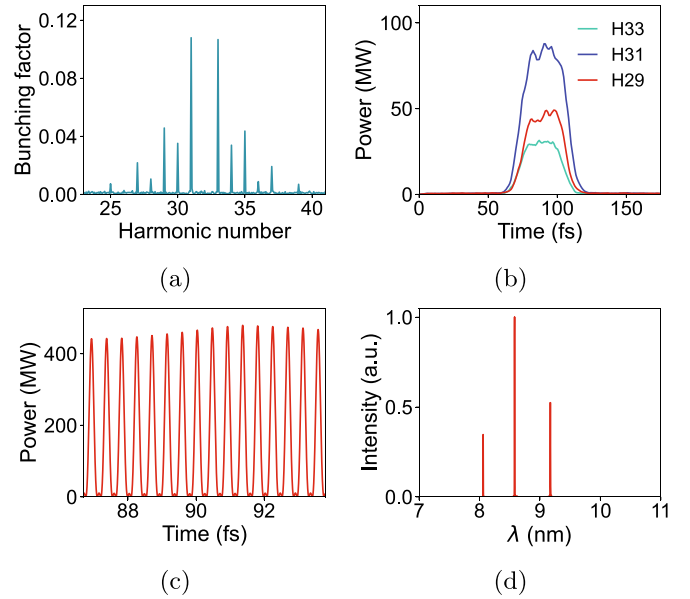


FIG. 10. High-harmonic bunching factor [panel (a)], radiation power of each harmonic [panel (b)], and the synthesized power [panel (c)] and spectrum [panel (d)] profile when shifting the spectrum one-harmonic order larger by varying B_1 from 8.6 to 8.9 in the EEHG scheme.

flexibility in waveform and spectrum control over the soft x-ray APTs' generation. The choice between consecutive and nonconsecutive harmonics is a trade-off driven by the specific temporal requirements of the target application (e.g., pulse duration versus repetition rate).

IV. DISCUSSION AND CONCLUSION

In general, we demonstrate a method that can generate soft X-ray APTs by combining the WS technique with the EEHG-FEL scheme. The waveform and spectrum of the APTs can also be controlled and adjusted easily taking advantage of the programmable high-harmonic bunching combs in the EEHG scheme. We want to emphasize that the fully coherent properties of each harmonic component in the synthesized APTs are preserved perfectly by the intrinsic seeded EEHG-FEL principle. However, the APTs' temporal contrast is apparently sensitive to the relative intensity distributions among the harmonic components. To assess how the relative intensity distribution among harmonic components affects the temporal contrast of synthesized APTs, we present a further analysis results in Fig. 11(a) based on the nonconsecutive harmonic synthesis (H_{32} : H_{30} : H_{28}). The relative intensities are set to be 1:0.5:1 (light blue curve), 1:1:1 (blue curve), 1:2:1 (yellow curve), and 1:4:1 (red curve), respectively. One can find out that increasing the relative weight of the central harmonic (H_{30}) will enhance the temporal contrast. When the intensity ratio is tuned to 1:4:1, the sidelobes are nearly suppressed to zero, yielding the highest signal-to-noise (SNR) ratio. However, this will also result in the pulse broadening in the main peak. If the central harmonic intensity is too high and weight exceeds this condition, the former sidelobe positions evolve into a quasicontinuous power pedestal, causing adjacent pulses in the train to merge and lose temporal

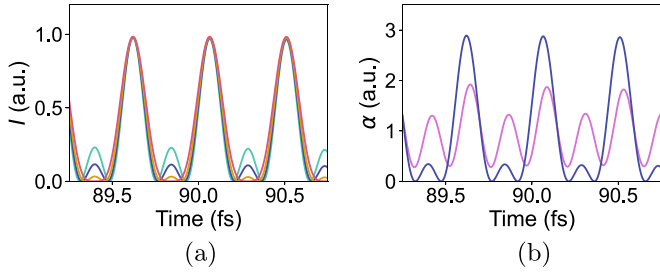


FIG. 11. The influences of the different relative intensity ratios [panel (a)] and the phase-matching conditions [panel (b)] on the synthesized APTs. In panel (a), the relative intensities are 1:0.5:1 (light blue curve), 1:1:1 (blue curve), 1:2:1 (yellow curve), and 1:4:1 (red curve), respectively. In panel (b), the blue curve is under the phase-matching condition while the magenta curve is not ($\Delta\Phi_{32,30} = -\pi/3$, $\Delta\Phi_{30,28} = \pi/5$).

isolation. In addition, an equal-intensity configuration (blue curve) will maximize the peak intensity enhancement factor $\alpha = (\sum M_k)^2 / \sum M_k^2$, in excellent agreement with the theoretical prediction in Sec. II C. Hence, the intensity ratio should be within the red and the blue curve to balance the peak intensity and the SNR of the synthesized APTs, which can be easily achieved through tuning the K of radiator segments. Furthermore, the linear phase-matching condition in Eq. (7) must be satisfied for the generation of synthesized APTs. We simulated the case where the relative phases of the three harmonics ($H_{32}:H_{30}:H_{28}$) deviate from the linear phase-matching condition specified in Eq. (7). Specifically, we introduced random phase offsets of $-\pi/3$ to H_{30} , H_{28} and $\pi/5$ to H_{30} , H_{32} . The result is shown in the magenta curve in Fig. 11(b). We can see the resulting waveform exhibits severe temporal degradation, in which a pronounced quasicontinuous power pedestal forms across the pulse trains, and the contrast between the main peak and adjacent sidelobes is drastically reduced. In contrast, in the blue curve in Fig. 11(b), the waveform synthesized under the matching condition maintains a clean temporal envelope with high peak-to-sidelobe contrast.

The practical applicability of the proposed scheme is subject to several intrinsic limitations, which are briefly discussed below. The cumulative energy-spread growth induced by multistage FEL amplification ultimately degrades high-harmonic microbunching, thereby imposing an upper limit on the number of harmonics that can be efficiently amplified and coherently synthesized. Moreover, the usable harmonic bandwidth is constrained by both the initial EEHG bunching spectrum and the finite, discretized longitudinal dispersion that can be introduced in practical radiator sections, which may prevent certain harmonic groups from reaching their optimal gain conditions. In addition, the quality of the synthesized waveform critically depends on the relative phase stability among different harmonic components, placing stringent requirements on seed-laser synchronization and phase-shifter control. Despite these intrinsic limitations, the requirements of the proposed scheme remain within the capabilities of modern high-performance seeded FEL facilities.

An additional point concerns the practical stability of the proposed scheme. In the present approach, harmonic generation originates from EEHG microbunching, whose stability

has been experimentally demonstrated under comparable operating conditions at facilities such as SXFEL and FERMI [44,45,50]. The subsequent harmonic evolution relies on the longitudinal dispersion provided by the undulator sections, which is intrinsically stable during routine operation. Moreover, the phase shifters employed here are permanent-magnet devices with fixed magnetic fields; the achievable gap adjustment precision of about $1\mu\text{m}$ corresponds to a phase accuracy on the order of 2mrad , which comfortably exceeds the phase stability requirements of the scheme. Taken together, these considerations indicate that the proposed multiharmonic synthesis approach is compatible with realistic operating conditions of modern seeded FEL facilities.

The experimental characterization of the proposed soft x-ray attosecond pulse trains also warrants brief discussion. Since attosecond waveform synthesis is only meaningful when accompanied by reliable temporal diagnostics, established attosecond metrology techniques are of direct relevance here. Methods such as RABBITT (Reconstruction of Attosecond Beating by Interference of Two-photon Transitions) [52] and CoBRA (correlation-based reconstruction of attosecond pulses) are appropriate for characterizing APTs. In particular, the correlation-based CoBRA technique, which is immune to delay jitter and enables robust retrieval of relative harmonic phases, is well matched to FEL conditions. CoBRA has been successfully extended to the soft x-ray regime and experimentally demonstrated at the FERMI [39]. These techniques provide a feasible experimental pathway to validate the attosecond pulse duration and waveform control enabled by the proposed scheme.

In conclusion, this work demonstrates a viable pathway for generating soft x-ray APTs with gigawatt peak power, sub-200 as pulse duration, and highly waveform and spectrum tunability. Theoretical analysis and three-dimensional numerical simulation results have been presented to validate the proposed method. Detailed optimization of five-harmonic synthesis gives out soft x-ray APTs with peak power of 3.5 GW and micropulse duration of 160 as. The nonconsecutive harmonic FEL lasing and synthesis, together with the shifts of the target harmonic components during the WS processes, demonstrate the flexible spectral and temporal control and adjustment of the synthesized APTs. The proposed method leverages precise control over harmonic components, relative phases, and radiator dispersions to shape the seeded FEL radiation pulses, thereby transforming the seeded FEL from a fully coherent single-harmonic source into a flexible platform for ultrafast laser pulse engineering. Beyond APTs' generation, the demonstrated capability to independently manipulate the relative phases and amplitudes of multiple harmonics offers a powerful degree of freedom for ultrafast science.

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available upon publication because it is not techni-

cally feasible and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

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- [1] M. F. Kling and M. J. J. Vrakking, Attosecond electron dynamics, *Annu. Rev. Phys. Chem.* **59**, 463 (2008).
 - [2] F. Calegari, D. Ayuso, A. Trabattoni, L. Belshaw, S. De Camillis, S. Anumula, F. Frassetto, L. Poletto, A. Palacios, P. Decleva, J. B. Greenwood, F. Martín, and M. Nisoli, Ultrafast electron dynamics in phenylalanine initiated by attosecond pulses, *Science* **346**, 336 (2014).
 - [3] D. Lu, M. A. Searles, and A. Klug, Crystal structure of a zinc-finger–RNA complex reveals two modes of molecular recognition, *Nature (London)* **426**, 96 (2003).
 - [4] M. Schultze, *et al.*, Delay in photoemission, *Science* **328**, 1658 (2010).
 - [5] Y. Morimoto and P. Baum, Diffraction and microscopy with attosecond electron pulse trains, *Nat. Phys.* **14**, 252 (2018).
 - [6] K. J. Schafer, M. B. Gaarde, A. Heinrich, J. Biegert, and U. Keller, Strong field quantum path control using attosecond pulse trains, *Phys. Rev. Lett.* **92**, 023003 (2004).
 - [7] P. Agostini, Nobel lecture: Genesis and applications of attosecond pulse trains, *Rev. Mod. Phys.* **96**, 030501 (2024).
 - [8] P. Antoine, A. L’Huillier, and M. Lewenstein, Attosecond pulse trains using high-order harmonics, *Phys. Rev. Lett.* **77**, 1234 (1996).
 - [9] P. M. Paul, E. S. Toma, P. Breger, G. Mullot, F. Augé, P. Balcou, H. G. Muller, and P. Agostini, Observation of a train of attosecond pulses from high harmonic generation, *Science* **292**, 1689 (2001).
 - [10] J. J. Macklin, J. D. Kmetec, and C. L. Gordon, High-order harmonic generation using intense femtosecond pulses, *Phys. Rev. Lett.* **70**, 766 (1993).
 - [11] M. Gebhardt and J. Limpert, Bright, high-repetition-rate water window soft x-ray source enabled by nonlinear pulse self-compression, *Light Sci. Appl.* **9**, 84 (2020).
 - [12] A. S. Johnson and J. P. Marangos, High-flux soft x-ray harmonic generation from ionization-shaped few-cycle laser pulses, *Sci. Adv.* **4**, eaar3761 (2018).
 - [13] S. M. Teichmann, F. Silva, S. L. Cousin, M. Hemmer, and J. Biegert, 0.5-keV soft x-ray attosecond continua, *Nat. Commun.* **7**, 11493 (2016).
 - [14] S. L. Cousin, F. Silva, S. Teichmann, M. Hemmer, B. Buades, and J. Biegert, High-flux table-top soft x-ray source driven by sub-2-cycle, CEP stable, 185- μ m 1-kHz pulses for carbon K-edge spectroscopy, *Opt. Lett.* **39**, 5383 (2014).
 - [15] J. Reinhard, F. Wiesner, T. Sidiropoulos, M. Hennecke, S. Kaleta, J. Späthe, J. J. Abel, M. Wünsche, G. Schmidl, J. Plentz, U. Hübner, K. Freiberg, J. Apell, S. Lippmann, M. Schnürer, S. Eisebitt, G. G. Paulus, and S. Fuchs, Soft X-ray imaging with coherence tomography in the water window spectral range using high-harmonic generation, *Light Sci. Appl.* **15**, 79 (2026).
 - [16] Y. Fu and E. J. Takahashi, High efficiency ultrafast water-window harmonic generation for single-shot soft x-ray spectroscopy, *Commun. Phys.* **3**, 92 (2020).
 - [17] T. Gaumnitz and H. J. Wörner, Streaking of 43-attosecond soft-x-ray pulses generated by a passively CEP-stable mid-infrared driver, *Opt. Express* **25**, 27506 (2017).
 - [18] P. K. Maroju, *et al.*, Attosecond pulse shaping using a seeded free-electron laser, *Nature (London)* **578**, 386 (2020).
 - [19] J. P. Duris, *et al.*, Controllable X-ray pulse trains from enhanced self-amplified spontaneous emission, *Phys. Rev. Lett.* **126**, 104802 (2021).
 - [20] S. Huang, Generating single-spike hard X-ray pulses with nonlinear bunch compression in free-electron lasers, *Phys. Rev. Lett.* **119**, 154801 (2017).
 - [21] J. Duris, *et al.*, Tunable isolated attosecond X-ray pulses with gigawatt peak power from a free-electron laser, *Nat. Photon.* **14**, 30 (2020).
 - [22] J. Yan, W. Qin, Y. Chen, W. Decking, P. Dijkstal, M. Guetg, I. Inoue, N. Kujala, S. Liu, T. Long, N. Mirian, and G. Geloni, Terawatt-attosecond hard X-ray free-electron laser at high repetition rate, *Nat. Photon.* **18**, 1293 (2024).
 - [23] Y. Xiao, C. Feng, and B. Liu, Generating isolated attosecond X-ray pulses by wavefront control in a seeded free-electron laser, *Ultrafast Sci.* **2022**, 9812478 (2022).
 - [24] Y. Xiao, T. Chen, B. Liu, Z. Huang, M. Pang, Y. Leng, and C. Feng, Isolated attosecond free-electron laser based on a subcycle driver from hollow capillary fibers, *Ultrafast Sci.* **5**, 99 (2025).
 - [25] S. Li, *et al.*, Attosecond coherent electron motion in Auger-Meitner decay, *Science* **375**, 285 (2022).
 - [26] S. Li, *et al.*, Attosecond-pump attosecond-probe x-ray spectroscopy of liquid water, *Science* **383**, 1118 (2024).
 - [27] T. Driver, *et al.*, Attosecond delays in X-ray molecular ionization, *Nature (London)* **632**, 762 (2024).
 - [28] A. M. Kondratenko and E. L. Saldin, Generating of coherent radiation by a relativistic electron beam in an undulator, *Part. Accel.* **10**, 207 (1980).
 - [29] R. Bonifacio, C. Pellegrini, and L. M. Narducci, Collective instabilities and high-gain regime in a free electron laser, *Opt. Commun.* **50**, 373 (1984).
 - [30] B. W. J. McNeil, N. Thompson, and D. Dunning, Transform-limited X-ray pulse generation from a high-brightness self-amplified spontaneous-emission free-electron laser, *Phys. Rev. Lett.* **110**, 134802 (2013).
 - [31] L. H. Yu, Generation of intense UV radiation by subharmonically seeded single-pass free-electron lasers, *Phys. Rev. A* **44**, 5178 (1991).
 - [32] L.-H. Yu, *et al.*, High-gain harmonic-generation free-electron laser, *Science* **289**, 932 (2000).
 - [33] G. Stupakov, Using the beam-echo effect for generation of short-wavelength radiation, *Phys. Rev. Lett.* **102**, 074801 (2009).
 - [34] D. Xiang and G. Stupakov, Echo-enabled harmonic generation free electron laser, *Phys. Rev. Spec. Top. Accel. Beams* **12**, 030702 (2009).

- [35] N. R. Thompson and B. W. J. McNeil, Mode locking in a free-electron laser amplifier, *Phys. Rev. Lett.* **100**, 203901 (2008).
- [36] C. Feng, J. Chen, and Z. Zhao, Generating stable attosecond x-ray pulse trains with a mode-locked seeded free-electron laser, *Phys. Rev. Spec. Top. Accel. Beams* **15**, 080703 (2012).
- [37] D. Xiang, Y. Ding, T. Raubenheimer, and J. Wu, Mode-locked multichromatic x rays in a seeded free-electron laser for single-shot x-ray spectroscopy, *Phys. Rev. Spec. Top. Accel. Beams* **15**, 050707 (2012).
- [38] D. J. Dunning, B. W. J. McNeil, and N. R. Thompson, Few-cycle pulse generation in an x-ray free-electron laser, *Phys. Rev. Lett.* **110**, 104801 (2013).
- [39] P. K. Maroju, C. Grazioli, M. Di Fraia, M. Moioli, D. Ertel, H. Ahmadi, O. Plekan, P. Finetti, E. Allaria, L. Giannessi, *et al.*, Complex attosecond waveform synthesis at FEL FERMI, *Appl. Sci.* **11**, 9791 (2021).
- [40] A. Wirth, M. Th. Hassan, I. Grguraš, J. Gagnon, A. Moulet, T. T. Luu, S. Pabst, R. Santra, Z. A. Alahmed, A. M. Azzeer, V. S. Yakovlev, V. Pervak, F. Krausz, and E. Goulielmakis, Synthesized light transients, *Science* **334**, 195 (2011).
- [41] B. Xue, K. Midorikawa, and E. J. Takahashi, Gigawatt-class, tabletop, isolated-attosecond-pulse light source, *Optica* **9**, 360 (2022).
- [42] B. Xue, Y. Tamaru, Y. Fu, H. Yuan, P. Lan, O. D. Mücke, A. Suda, K. Midorikawa, and E. J. Takahashi, A custom-tailored multi-TW optical electric field for gigawatt soft-X-ray isolated attosecond pulses, *Ultrafast Sci.* **2021**, 9828026 (2021).
- [43] P. K. Maroju, *et al.*, Attosecond temporal structure of non-consecutive harmonic combs revealed by multiple near-infrared photon transitions in two-color photoionisation, *Commun. Phys.* **8**, 207 (2025).
- [44] P. Rebernik Ribič, A. Abrami, L. Badano, M. Bossi, H.-H. Braun, N. Bruchon, F. Capotondi, D. Castronovo, M. Cautero, P. Cinquegrana, *et al.*, Coherent soft x-ray pulses from an echo-enabled harmonic generation free-electron laser, *Nat. Photon.* **13**, 555 (2019).
- [45] C. Feng, *et al.*, Coherent and ultrashort soft x-ray pulses from echo-enabled harmonic cascade free-electron lasers, *Optica* **9**, 785 (2022).
- [46] C. H. Shim, H. Yang, J. Hong, G. Kim, H.-S. Kang, and M. H. Cho, Intensity optimization of x-ray free-electron laser by using phase shifters, *Phys. Rev. Accel. Beams* **23**, 090702 (2020).
- [47] K.-J. Kim, Z. Huang, and R. Lindberg, *Synchrotron Radiation and Free-electron Lasers: Principles of Coherent X-ray Generation* (Cambridge University Press, Cambridge, 2017).
- [48] L. H. Yu and J. Wu, Theory of high gain harmonic generation: An analytical estimate, *Nucl. Instrum. Methods Phys. Res. Sect. A* **483**, 493 (2002).
- [49] E. Prat, W. Hu, C. Arrell, M. Calvi, P. Dijkstal, R. Follath, and S. Reiche, Experimental demonstration of mode-coupled and high-brightness self-amplified spontaneous emission in an X-ray free-electron laser, *Phys. Rev. Lett.* **133**, 205001 (2024).
- [50] B. Liu, C. Feng, D. Gu, F. Gao, H. Deng, M. Zhang, S. Sun, S. Chen, W. Zhang, W. Fang, Z. Wang, Q. Zhou, Y. Leng, M. Gu, L. Yin, Q. Gu, G. Fang, D. Wang, and Z. Zhao, The SXFEL upgrade: From test facility to user facility, *Appl. Sci.* **12**, 176 (2021).
- [51] S. Reiche, Genesis 1.3: A fully 3D time-dependent FEL simulation code, *Nucl. Instrum. Methods Phys. Res. Sect. A* **429**, 243 (1999).
- [52] L. Cattaneo, J. Vos, M. Lucchini, L. Gallmann, C. Cirelli, and U. Keller, Comparison of attosecond streaking and RABBITT, *Opt. Express* **24**, 29060 (2016).