

Single-pass synchronized two-color attosecond x-ray free-electron laser featuring attosecond time jitter

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(Received 8 December 2025; accepted 25 March 2026; published 15 April 2026)

Two-color attosecond x-ray pulses are the key to advancing the study of ultrafast dynamics in quantum systems. In this work, we propose a single-pass free-electron laser scheme generating synchronized attosecond two-color x-ray pulses with femtosecond separation and tens-of-gigawatts peak power. By modulating a chirped electron beam with an infrared laser, we create a periodic train of ultrashort current spikes. This scheme utilizes the limited gain bandwidth of the free-electron laser as a filter, allowing only two adjacent current spikes with different resonance wavelengths to lase. This scheme holds promise for delivering sub-300-as two-color x-ray pulses with an intrinsic timing jitter below 100 as. This work provides insights and technical support for generating two-color attosecond x-ray free-electron laser pulses with robust time jitter.

DOI: [10.1103/cw2t-yvwd](https://doi.org/10.1103/cw2t-yvwd)

I. INTRODUCTION

In the microscopic world, dynamic phenomena exhibit intrinsic ultrafast characteristics, and different physical processes occur on different timescales. For instance, atomic and molecular dynamics, as well as lattice motions, take place on the femtosecond timescale, while the ultrafast dynamics of electrons occur on the attosecond timescale [1]. Pump-probe spectroscopy serves as the primary experimental methodology for exploring ultrafast dynamic processes [2]. In this experimental setup, an initial ultrashort pulse is utilized to excite the system into a nonequilibrium state, after which a subsequent ultrashort pulse serves to probe the transient evolution of the system's state. The minimization of time jitter is also critically important in pump-probe experiments, as it directly dictates the achievable temporal resolution and the fundamental integrity of the measurement. Therefore, the development of stable ultrashort pump-probe techniques, particularly stable attosecond pump probe, is of paramount importance. The experimental demonstration of isolated attosecond pulse production through high-harmonic generation (HHG) was first achieved in 2001 [3]. HHG light sources have produced two-color attosecond pulse pairs in the extreme-ultraviolet regime

[4,5], which makes it possible to implement attosecond pump-probe experiments. Although HHG light sources have been extended to the soft x-ray regime [6–8], it still poses a big challenge due to the notably low efficiency of high harmonic conversion. Presently, attosecond science research, such as the dynamics of inner-shell electrons in atoms, urgently requires the development of stable attosecond x-ray pump-probe technology.

Over the past two decades, x-ray free-electron lasers (XFELs) have undergone rapid development, and now there are several XFEL user facilities in operation around the world [9–14]. This progress has enabled the generation of ultrashort x-ray two-color pulses and has provided great opportunities for conducting x-ray pump-probe experiments with time resolution ranging from femtosecond to attosecond. XFEL radiation arises from relativistic electron bunch going through an undulator [15]. Compared with HHG light sources, XFEL possesses higher energy conversion efficiency in the x-ray regime, typically around 0.1%. For example, a typical electron beam with terawatt peak power can generate an x-ray pulse with a peak power of tens of gigawatts [16], several orders of magnitude higher than HHG light sources [7]. The resonance wavelength for FELs is given by the equation $\lambda_r = \lambda_u(1 + K^2/2)/2\gamma^2$, where λ_u , K , and γ are the undulator period, the undulator magnetic field strength parameter, and the Lorentz factor of the electron beam, respectively. In FEL facilities, the undulator period is fixed, and the tunable degrees of freedom for radiation wavelength are exclusively K and γ . Therefore, there are two general approaches for generating two-color FEL pulses. The first approach involves passing an electron beam sequentially through two undulators with different magnetic field strengths [17–31], while the second approach delivers electrons of different energies through the

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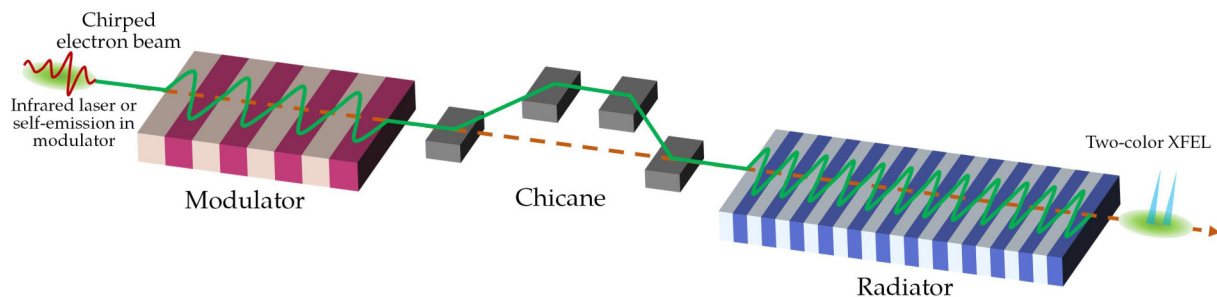


FIG. 1. Layout of the attosecond two-color setup. The modulator introduces energy modulation to the large energy chirped electron beam. The chicane compresses the beam to generate multiple current peaks. In the radiator, the radiation from the majority of current spikes is suppressed, with the exception of two adjacent current spikes around the resonant condition, which results in attosecond two-color XFEL pulse generation.

same undulator [32–40]. In the following, we provide a brief overview of two-color FEL schemes.

For the first two-color FEL generation approach, two-color FEL operation was first demonstrated in the midinfrared region [17], and subsequently, the generation of two-color FEL pulses was achieved in ultraviolet [18] and x-ray regimes [19–21]. In this configuration, the peak power of the two FEL pulses is limited because the same electron beam is utilized to produce FEL radiation in both stages. The peak power can be improved by permitting only a portion of the beam to lase in each undulator section, such as electron beam tilt-based fresh-slice FEL [21–24] and slice matching-based fresh-slice FEL [25–27]. Additionally, more two-color FEL schemes, based on this approach, have been proposed and demonstrated for controlling polarization [28,29] and minimizing duration to attosecond [30,31].

For the second two-color FEL generation approach, it was first demonstrated with twin electron bunches at different energies [32,33]. There have also been some schemes proposed based on chirped electron beams [34–40]. These schemes generate two-color FEL pulses by creating local current spikes and suppressing lasing in other parts, such as the emittance spoiling strategy [34,35], the temporally shaped laser heater technique [36], the sextupole magnet-based scheme [37], the photocathode laser emittance spoiler method [38], the wakefield excitation-based scheme [39], and the nonlinear compression [40]. Supplementarily, there are another two-color FEL solutions that are not so straightforward to classify into the above two approaches, such as the chirped seed scheme [41,42], the seeded FEL based on tilted beam lasing in transverse gradient undulator [43], the single-crystal self-seeded FEL with chirped electron beam [44], the split-and-delay line based on multilayer Bragg mirrors [45], and the Laue x-ray compressor method [46]. There are also schemes that utilize two seed pulses with different wavelengths and a temporal separation, such as the pulses are copropagated with the same electron bunch through a single undulator [47] or through two undulator sections that are individually tuned to the respective seed wavelengths [48]. These findings indicate a promising future for the generation of ultrashort two-color pulses based on FELs, paving the way for ultrafast dynamic phenomena investigations.

In this work, we propose a single-pass scheme for generating synchronized two-color attosecond FEL pulses. The

proposed scheme features attosecond-level timing jitter. As illustrated in Fig. 1, this scheme employs an infrared laser to modulate an electron beam with a large energy chirp or, alternatively, achieves the modulation by using self-modulation of the infrared radiation emitted by the tail of the electron beam [30,49]. Then, the modulated electron beam passes through a magnetic chicane to produce multiple current spikes. Finally, in the radiator, only the x-rays emitted by the two adjacent central current spikes can obtain sufficient gain due to the limited gain bandwidth of FEL, resulting in the generation of attosecond two-color XFEL pulses. The two current spikes, generated by infrared laser modulation and subsequent chicane compression, exhibit extremely stable temporal separation. This inherent stability is the direct physical mechanism that enables attosecond-level timing jitter between the resulting two-color XFEL pulses. Here, we provide an explanation regarding the limited gain bandwidth: When the electron beam chirp is increased to broaden the FEL bandwidth, the bandwidth of the radiation does not increase indefinitely due to factors such as increased emittance, poorer alignment, and stronger mismatch. Instead, it reaches a limit, which is what we refer to as the limited gain bandwidth.

This paper is organized as follows. In Sec. II, we present numerical simulations to validate the proposed scheme. We first simulate the phase space and current distribution of the electron beam before and after modulation and compression. This is followed by an analysis of the process in which the modulated beam radiates in the undulator to generate two-color XFEL pulses. In Secs. III and IV, we provide a discussion that compares the advantages of our scheme against other typical two-color XFEL schemes and concludes with a brief summary of this work.

II. NUMERICAL VALIDATION

To better understand this scheme, we illustrate it through numerical simulation, and the calculation is performed by using GENESIS 1.3 [50]. The simulation parameters are summarized in Table I.

For the simulation framework, we adopted the parameters of Shanghai High Repetition Rate XFEL and Extreme Light Facility (SHINE). The large energy chirp in electron bunches can be generated either by modulation with high-power infrared lasers or by accelerating the electron bunch

TABLE I. The relevant parameters of FEL simulation.

Parameter	Value	Unit
Electron energy	8.0	GeV
Energy spread	0.8	MeV
Peak current	1500	A
Photon energy E_0	10	keV
Undulator period λ_u	2.6	cm
Undulator length in unit of λ_u	152	—
Drift length in unit of λ_u	40	—
Number of undulator segments	24	—
Electron bunch length	6.0	μm
Average beta function	23	m
Normalized emittance	0.4	mm mrad
Undulator parameter K_1	1.632	—
Modulator period	40	cm
Modulator parameter K_2	49.5	—

in the off-peak phase of radio frequency structures [51]. In the simulation, the electron beam energy is 8 GeV and the energy chirp of the electron beam is around 9.6 MeV/fs, which is produced by the modulation of a high-power CO₂ laser in a wiggler, as shown in Fig. 2(a). Then, the chirped electron bunch is modulated by an infrared laser with wavelength of 1 μm in the modulator. The period of the modulator is 40 cm, the energy modulation depth is 8, the length of the modulator is 2.4 m, the peak power of the infrared laser is 10 GW, and the resonant wavelength in the modulator is 1 μm . Here, the modulation depth is defined in units of the rms energy spread of the electron beam before modulation. The period of the radiator is 2.6 cm, and the resonant photon energy in the radiator is around 10 keV. The phase space distributions of the electron beam before and after the modulator are depicted in Figs. 2(a) and 2(b), respectively. Meanwhile, the phase space and current profile of the electron beam after the chicane are shown in Fig. 2(c). The simulation results indicate that the modulated electron beam forms multiple ultrashort current spikes after passing through the chicane. In the subsequent radiation process within the undulator, limited by the FEL gain bandwidth, only two of these current spikes experience sufficient gain to lase, thereby generating two-color attosecond XFEL pulses. The value of R_{56} adopted here is 0.15 mm, and the length of the chicane is 2 m.

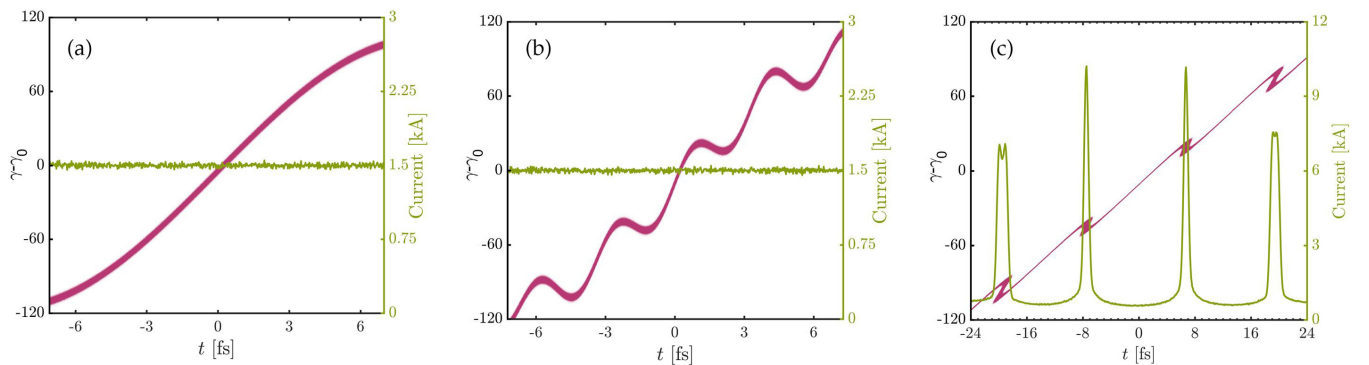


FIG. 2. (a) The phase space and current distribution of the electron beam before the modulator. (b) The phase space and current distribution of the electron beam after the modulator. (c) The phase space and current distribution of the electron beam after the chicane.

Figure 3 presents the simulation results of the electron beam generating two-color attosecond pulses in the radiator. The evolution of the normalized power distribution and normalized spectrum distribution in the radiator is presented in Figs. 3(a) and 3(b), respectively. Although all ultrashort current spikes emit radiation during the startup process, only the radiation generated by the two current peaks in the core of the electron beam experiences sufficient FEL gain to evolve into two-color attosecond XFEL pulses. The evolution of the pulse duration and bandwidth of the two-color XFEL pulses in the radiator is shown in Figs. 3(c) and 3(d). It can be found that the bandwidth of the generated two-color attosecond XFEL pulses gradually decreases during the gain process and stabilizes after saturation. Similarly, the pulse duration also reaches a stable state after saturation. The power and spectral distributions of the two-color attosecond XFEL radiation around the saturation point (at $z = 80$ m) are shown in Figs. 3(g) and 3(h), respectively. At the saturation point, the full widths at half maximum (FWHM) of the pulse durations of the two pulses are 338 ± 31.6 and 320 ± 31.2 as, respectively. The FWHM of the bandwidths are 7.9 ± 0.7 and 8.9 ± 0.8 eV, respectively. Both pulses have peak average powers exceeding 4 GW, and their time-bandwidth products are approximately 1.52 (1.56) times the Fourier transform limit. The timing jitter between the two pulses is 90 as (rms). It should be noted that the time delay between the two-color pulses is not the 3.3 fs corresponding to the modulation period, but instead becomes 14.2 fs. This is due to the stretching caused by the R_{56} of the chicane. After saturation, there is a slight increase in both pulse duration and bandwidth, but they remain essentially stable. This is due to the slippage effect and changes in the resonance relationship caused by the reduction in electron beam energy. At the exit of the radiator, the average pulse durations and bandwidths of the two-color pulses are approximately 360 as and 12 eV, respectively. The time-bandwidth products approximately increase to 2.4 times the Fourier transform limit. The evolution of the pulse energy and peak power of the two-color XFEL pulses in the radiator is shown in Figs. 3(e) and 3(f), respectively. At the exit of the radiator, the average energy and average peak power of the two-color XFEL pulses are approximately 4 μJ and 10 GW, respectively. The timing jitter between the two pulses is 117 as (rms). It should be pointed that in the above FEL simulations, we did not taper the radiator. Therefore, it is expected

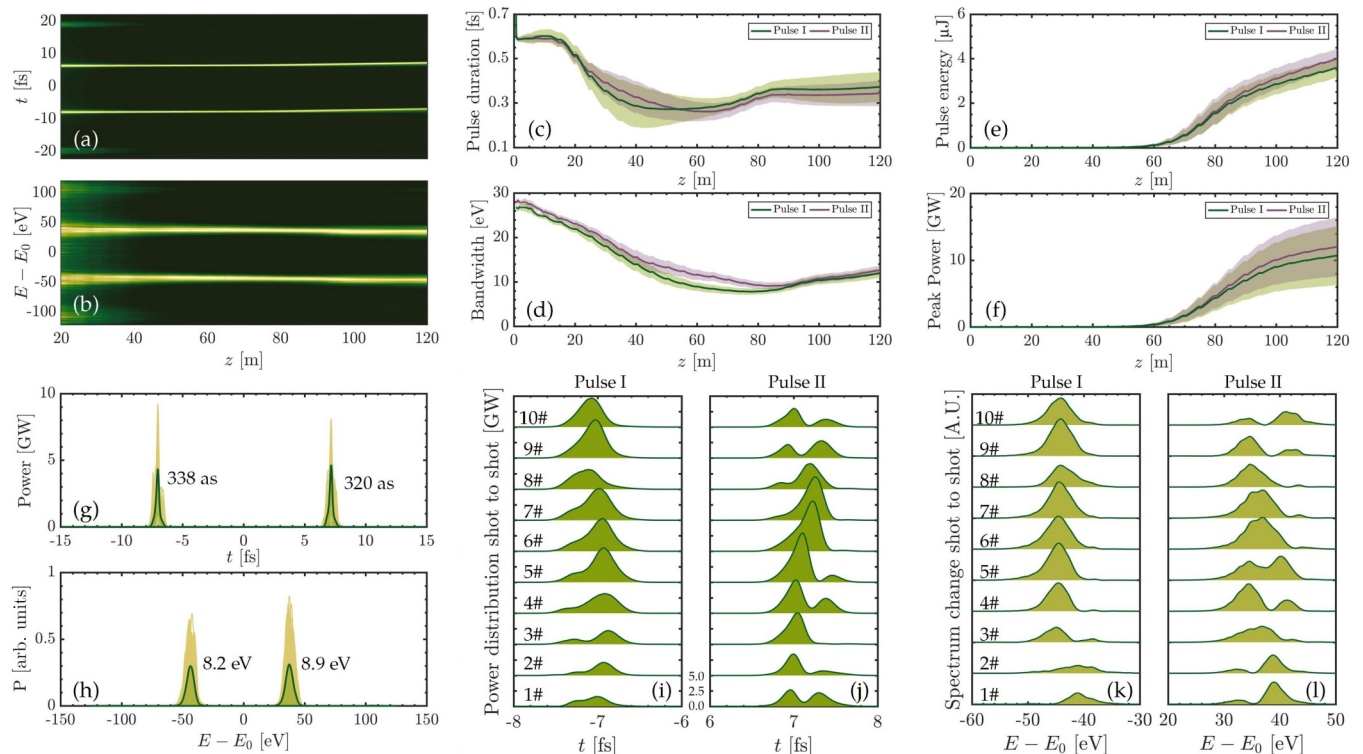


FIG. 3. (a), (b) Evolution of the normalized power and normalized spectrum of attosecond two-color XFEL pulses in the radiator. (c), (d) Evolution of attosecond two-color XFEL pulse duration and spectral bandwidth in the radiator. The shaded band indicates the $\pm\sigma$ (rms) confidence interval around the mean value. (e), (f) Evolution of attosecond two-color XFEL pulse energy and peak power in the radiator. The shaded band indicates the $\pm\sigma$ (rms) confidence interval around the mean value. Panels (g) and (h) show the power and spectrum distribution of 100-shot numerical simulations near the saturation point, where the green curve indicates the averaged power distribution and the yellow curve indicates single-shot power distribution. Panels (i)–(l) show the power and spectrum distribution of ten simulations near the saturation point. Pulse I and Pulse II in the figure refer to the first and second pulses of the two-color XFEL pulses, respectively.

that tapering the radiator would significantly boost the pulse energy and peak power, and help mitigate the increase in the time-bandwidth product to some extent.

In the numerical simulations, we only varied the seed values in GENESIS. The results indicate that the best achievable timing jitter between the generated two-color pulses is around 100 as. The primary focus of this work is to demonstrate the feasibility of this scheme in principle, and a flat-top current profile was adopted. There are other potential factors that influence the timing jitter, such as jitter in R_{56} and electron beam energy chirp. For instance, jitter in the chicane R_{56} also contributes to timing jitter: A 0.01% jitter in R_{56} would increase the timing jitter from 100 to 112 as. For the jitter induced by electron beam energy chirp, in this scheme, the interval between adjacent current spikes is 3.3 fs. We can readily evaluate that to achieve a time jitter of less than 50 as, the tolerable energy chirp jitter is 1.5%, which provides a substantial margin and is experimentally achievable. In the experiment by Lutman *et al.*, the electron beam energy chirp jitter was less than 0.2% [19]. Under current technical conditions, the timing jitter between the two-color pulses caused by jitter in R_{56} of the chicane and the electron beam energy chirp can potentially be reduced to a negligible level.

It is well known that conventional self-amplified spontaneous emission (SASE) pulse exhibits multiple modes in

longitudinal dimension, corresponding to multiple spikes in the spectrum. In this scheme, the produced two-color attosecond XFEL pulses around saturation point ($z = 80$ m), compared with conventional SASE pulses, are predominantly characterized by single-peak and double-peak profiles in the longitudinal distributions illustrated in Figs. 3(i) and 3(j), and the corresponding spectral distributions are presented in Figs. 3(k) and 3(l). According to data statistics, the proportions of single-peak and double-peak pulses are 76% and 24%, respectively. It should be pointed out that the predominance of single-peak and double-peak pulses is attributed to the fact that the pulse length is approximately twice the FEL cooperation length.

In the simulation, the gain bandwidth of x-ray radiation is about 130 eV (FWHM), which is corresponding to a time window of 5.4 fs. Combined with an electron beam chirp of 9.6 MeV/fs and a modulation wavelength of 1 μ m, it can be calculated that the jitter between the modulating laser and the electron beam needs to be less than 0.5 fs (rms), which is corresponding to a phase jitter of $\pi/8$ (rms). This brings a great challenge to this scheme. However, there is no need to be discouraged. The self-modulation of the electron beam is capable of producing stable modulation [49], and the phase jitter reduces down to $\pi/15$ (rms) at the modulation period of ~ 3 μ m. By using the self-modulation strategy, the stability can be improved dramatically. Therefore, it is

still promising for the experimental implementation of our scheme.

III. DISCUSSION

In pump-probe experiments, achieving extremely high temporal delay precision is a fundamental prerequisite for capturing ultrafast dynamics. Currently, although light sources such as XFELs can generate ultrashort pulses from femtosecond to attosecond scale, the time jitter of tens to hundreds of femtoseconds between the pump laser pulse and the probing XFEL pulse severely limits the ultimate resolution of experiments [12,52–56]. This timing uncertainty renders any single-shot measurement unreliable, compelling experiments to rely on extensive repetition and signal averaging to obtain meaningful data. Therefore, reducing temporal jitter is of critical importance: first, it directly enhances temporal resolution, enabling precise measurements of physical, chemical, or biological dynamics evolving on femtosecond or even attosecond timescales. Second, it significantly improves the signal-to-noise ratio and experimental efficiency; smaller jitter means that data acquired in each shot are more faithful, thereby substantially reducing the number of required experimental repetitions. Ultimately, it will advance the experimental paradigm from the current “statistical averaging” toward “true single-shot measurements” [55,56], which is crucial for studying irreversible or fluctuating transient phenomena. That is, the control of time jitter serves as the key bridge connecting the capability to generate ultrashort pulses with the measurement of ultrahigh temporal resolution, directly determining the depth of our exploration into the microscopic dynamics of matter.

Therefore, in the generation of attosecond two-color XFEL, the time jitter between the two-color pulses is critical. For two-color XFEL schemes based on the two-bunch mode, the jitter between pulses originates from the relative timing jitter between electron bunches, typically on the order of tens to hundreds of femtoseconds [55,56]. For fresh-slice-based two-color XFEL schemes, the temporal jitter between the two-color pulses is on the order of a few femtoseconds [22,26]. For two-color schemes based on x-ray laser-enhanced attosecond pulse generation (XLEAP), the temporal jitter between the two-color pulses is on the order of several hundred attoseconds [31]. Our proposed two-color XFEL scheme holds the potential to reduce the temporal jitter between the two-color pulses to below 100 as.

Among the various two-color XFEL schemes, the attosecond two-color XFEL approach based on the two-bunch method suffers from the drawback of limited and difficult control over the temporal delay between the two pulses [32,33]. The fresh-slice-based two-color XFEL scheme requires a longer undulator line and faces challenges in reducing the pulse duration to attosecond [21–27]. The XLEAP-based two-color XFEL scheme offers a narrow range of adjustability for the delay between the generated attosecond two-color pulses [30,31]. As for the single-crystal self-seeding two-color XFEL scheme, inherent crystal-induced stretching makes it difficult to produce attosecond two-color pulses, while the photon energy tunability of the two-color XFEL pulses is also constrained [44]. In the proposed attosecond two-color XFEL

scheme, the temporal delay between the two-color pulses is jointly determined by the electron bunch chirp, modulation depth, and modulation wavelength, enabling delays ranging from a few to tens of femtoseconds. While the proposed scheme has the potential to generate two-color pulses with excellent synchronization, it cannot achieve zero delay adjustment between the pulses. To a certain degree, the proposed scheme and the XLEAP two-color scheme exhibit complementarity in the scope of their temporal delay adjustability. In terms of how time delay tunability is achieved, unlike other schemes that predominantly rely on the chicane for delay control, this scheme enables delay adjustment by combining the chicane with variations in the modulation laser power (adjusting the modulation depth). The pulse duration of this scheme can be controlled by adjusting the wavelength of the modulation laser. For instance, reducing the modulation laser wavelength can significantly shorten the generated attosecond two-color XFEL pulses.

IV. SUMMARY

In conclusion, the innovation of this work lies in utilizing the FEL gain bandwidth as a natural filter to generate synchronized two-color attosecond x-ray pulses. This is achieved by precisely controlling two current spikes that lase simultaneously within the FEL gain bandwidth, thereby eliminating the need for magnetic chicane between undulators. Since the two-color pulses originate from two adjacent spikes of the same periodically modulated chirped electron beam, the proposed scheme ensures highly stable temporal synchronization between them, with timing jitter potentially reduced to the 100 as level. Numerical simulations show that around 10 keV, two-color XFEL pulses with a peak power of 10 GW, a pulse length of ~ 360 as, a temporal separation of 14.2 fs, a time jitter around 100 as, and $\sim 1\%$ energy separation can be produced. The pulse duration can be effectively reduced by decreasing the wavelength of the modulation laser. The performance of this scheme in terms of peak power can be further enhanced by increasing the peak current and using undulator tapering strategy. This scheme is flexible and can be tailored to meet the requirements of different radiation photon energies and delays between two-color pulses, and optimized for the parameters of different XFEL user facilities.

ACKNOWLEDGMENTS

This research was funded by the Guangdong Basic and Applied Basic Research Foundation (2026A1515010336), the National Natural Science Foundation of China (12575342, 22288201, 12005135), the DICP (DICP I202304) and the Strategic Priority Research Program of the Chinese Academy of Sciences [XDB0970000 (subject: XBD0970100)].

DATA AVAILABILITY

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