A perfect X-ray beam splitter and its applications to time-domain interferometry and quantum optics exploiting free-electron lasers

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Abstract

Generation of Phase-Locked X-Ray Pulses

Our approach to produce phase-locked pulses in the soft and hard X-ray regime is to overlay a fully coherent signal on an electron bunch that only lases in two well-defined longitudinal slices. It is achieved with a method (Fig. 1) that combines a “slotted” foil (11, 12) (or, equivalently, energy modulation or laser slicing, which may be more appropriate for high-repetition-rate FELs), transverse tilting of the electron beam (13–15), and self-seeding (16–20). The latter is a two-stage process to generate coherent X-ray pulses: The first FEL stage consists of a standard self-amplified spontaneous emission (SASE) section, whose output is spectrally filtered by a monochromator. This signal is then passed through a phase-locked, high-energy photon pulse, which is delayed with respect to the SASE output by a fixed amount. The second FEL stage is then triggered by this delayed photon pulse, resulting in a photon emission scheme. Instead of splitting the photon pulses after their generation by the FEL, we split the electron bunch in the accelerator, prior to photon generation, to obtain phase-locked X-ray pulses with subfemtosecond duration. Time-domain interferometry becomes possible, enabling the concomitant program of classical and quantum optics experiments with X-rays. The scheme leads to scientific benefits of cutting-edge FELs with attosecond and/or high-repetition rate capabilities, ranging from the X-ray analog of Fourier transform infrared spectroscopy to damage-free measurements.

Significance

Brilliant, ultrashort, and coherent X-ray free-electron laser (FEL) pulses allow investigations of dynamics at the inherent time and length scale of atoms. However, missing are sequences of phase-locked X-ray pulses, desirable for time-domain correlation spectroscopies and coherent quantum control. Based on selective electron-bunch degradation in the accelerator, combined with two-stage, self-seeded photon emission, we propose an FEL mode, generating subfemtosecond, phase-locked X-ray pulse pairs with up to 100 fs delay. Splitting the electron bunch in the accelerator, instead of photon pulses in the beamline, avoids relative phase jitter. This enables time-domain interferometry, such as the X-ray analog of the ubiquitous Fourier transform infrared spectrometer, and, more generally, all of nonlinear and quantum optics requiring coherent copies of beams. The Future of Accelerators, A. W. Chao, W. Chou, Eds. (2019), vol. 10, pp. 33–48

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Movable in vertical direction

Fig. 1. (A) X-ray FEL beamline layout with a movable microfabricated mask (blue) and higher-order multipole magnets (purple) in a dispersive section, as well as the two undulator sections (pink–green) that are separated by a self-seeding chicane (gray–green). (B) The movable mask features a set of slots: One set preserves the electron bunch (pink) for SASE generation, whereas two narrow slits define the unspoiled parts of the electron bunch, from which the coherent signal originates. (C) The electron bunch is shaped with nonlinear transverse tilts using multipole magnets and realigned in the undulator section: For short (long) time delays, it is aligned on axis with the tails (central part) in the first and the central part (tails) in the second stage.

then used as a “seed” for the second stage and overlapped with the “sliced” parts of the electron bunch. In our method, the electron beam contains two parts defined by the slotted foil: One part is unspoiled, while the other part is spoiled, except for the two regions, as defined by the slits on a microfabricated mask (Fig. 1B). We separate the two regions transversely and correct the global beam trajectory such that one region is aligned to the SASE and the other region to the self-seeding stage. As a result, the first part drives the SASE section in saturation, while the two unspoiled slices in the second stage amplify the seed signal to produce a phase-locked pulse pair.

The required spatial separation of the two regions is achieved by imposing a transverse tilt on the electron bunch in the dispersive section (see Fig. 1C and the start-to-end simulations in SI Appendix). Thereby, in both FEL stages, either the central part or the tails of the bunch is/are aligned to the undulator axis, whereas the respective other part undergoes betatron oscillations and does not contribute to the lasing process. The beam tilt is imposed where also the microfabricated mask is placed. Multipole magnets in the dispersive section can be used to alter the longitudinal and transverse position of the electron bunch, which is then preserved downstream. Namely, quadrupole magnets result in linear, sextupoles in quadratic, octupoles in cubic displacements, etc. A combination of a sextupole and a decapole magnet yields the desired step-function profile indicated in Fig. 1C. To switch between short and long time delays among the phase-locked pulses, the mask is moved vertically, and the alignment of the electron tilt is flipped.

Fig. 2 shows the performance of the PHLUX mode, in terms of radiation profile and spectrum, as well as phase stability, given the baseline capabilities of the SwissFEL Athos soft X-ray branch (21), assuming a seed energy of $E \sim 1.097 \text{eV}$ and a self-seeding chicane with a resolving power of 50,000, foreseen as a future upgrade of the beamline. Here, the PHLUX mode is benchmarked in the soft X-ray regime, but the concept is equally applicable to hard X-rays. To create a phase-stable wave train, the bandwidth of the monochromators must be significantly smaller than the spectral width (inverse length) of individual SASE spikes (see discussion on self-seeding in SI Appendix). The modal structure in the spectrum depends on the temporal separation of the slices and, at the maximum $\Delta t_{\text{max}} = 96 \text{fs}$ considered here, corresponds to an energy resolution of $1/\Delta t_{\text{max}} \sim 45 \text{meV}$, assuming Fourier-limited pulses; a smaller self-seeding resolving power, such as $\sim 5,000$ implemented at Linac Coherent Light Source (LCLS) (19), would still give a useful $\Delta t_{\text{max}} \lesssim 20 \text{fs}$. A peak radiation power of $\sim 2 \text{GW}$ results in a peak photon field strength of $\sim 1 \text{MV/cm}$ at the source point. To reach nonlinear driving regimes, this field strength can be further increased; e.g., the baseline focusing capabilities of SwissFEL Athos allow for an increase by a factor of $\sim 50$ at the sample position.

The total width of the spectrum, i.e., the number of modes, depends on the slit width on the mask—wider slits result in narrower spectra. PHLUX delivers a minimal pulse separation of $\Delta t \sim 2 \text{fs}$, which is limited by the transverse size of the electron beam. In turn, it also determines the slit width of the mask, i.e., the minimal pulse duration. We note that 2 fs is the full-width at half-maximum (FWHM) of the electron slice, whereas lasing occurs mainly from the central portion, yielding a shorter photon pulse length of 0.5 fs FWHM. Our scheme is rather insensitive to the accuracy of the microfabrication process: Realistic tolerances, of order $\mu \text{m}$, result in pulse length changes of less than...
100 as and have negligible effect on the phase and amplitude relation of the pulses.

**Stability of Phase-Locked X-Ray Pulses**

Because the two pulses follow a common path, the PHLUX scheme is fundamentally different and offers much higher phase stability than conventional X-ray split-and-delay approaches (22–24), where slight vibrations in the delaying monochromators and mirrors translate into phase jitter between the two pulses: For a central photon energy of $E \sim 1.097$ eV, $\Delta \phi = 1$ rad corresponds to a path-length difference of only $\sim 1.8$ Å. This translates into stability unachievable for standard setups, typically consisting of eight optical elements, which need to be set and held in place with respect to each other. In contrast, Fig. 2B shows that the peak-to-peak phase stability of PHLUX is always much better than $\Delta \phi \sim 60$ mrad. The absolute phase $\phi$ is not controllable, but, importantly, jitter in $\Delta t$ and $\phi$, determined by the mask stability and quality of the self-seeding monochromator, does not affect $\Delta \phi$ (see discussion on sources of beam jitter in Materials and Methods). That is, the FEL itself is used as a “perfect” beam splitter, which protects the phase difference $\Delta \phi$ from noticeable jitter. The clean, two-slit interference fringes in the radiation power spectrum (Fig. 2C), corresponding to the delay between pulses, attest to the quality of the pulse replication.

The phase stability of pulse pairs can be expressed by the first-order correlation function, generally defined as

$$g^{(1)}(t_1, t_2) = \frac{\langle E(t_1)E^*(t_2) \rangle}{\langle |E(t_1)|^2 \rangle \langle |E(t_2)|^2 \rangle}.$$  

$E(t)$ is the radiation field, which is evaluated at the times $t_1$ and $t_2 = t_1 + \Delta t$ of the peak of the first and second pulse, respectively. $\Delta t$ is the pulse separation set by the machine parameters and the position of the slotted foil in the electron beam. Due to the phase rigidity of the individual pulses, there is no need for evaluation of a further time integral. An average is taken over many shots. To calculate $g^{(1)}$, we consider that the starting signal arises due to shot-noise of the incoherent spontaneous radiation from the electron beam. Importantly, neither SASE amplification nor the self-seeding monochromators change the white-noise characteristic of this starting signal (25). Therefore, the coherence properties are defined by the resolving power of the monochromators, and a Monte Carlo evaluation permits us to determine the coherence function $g^{(1)}(t_1, t_2)$. Amplification of two slices of the output field of the monochromators then inherits these coherence properties, unless the pulses are driven deep into saturation, which anyhow should be avoided, as it distorts the spectral quality of the FEL signal. Due to the nature of white noise, a fixed phase relation also results in a stable amplitude relation (26).

Fig. 3 illustrates the relative phase stability as a function of $\Delta t = t_2 - t_1$, reconstructed via $g^{(1)}(t_1, t_2)$. The signal degrades with increasing $\Delta t$, indicating the importance of a self-seeding chicane with highest possible resolving power. $g^{(1)} = 1$ means that the relative phase difference between two pulses is stable over many shots (coherent light), while for $g^{(1)} = 0$, the relative phase between pulses fluctuates randomly (chaotic light). A source for the reduced stability at large $\Delta t$ could, in principle, be intrinsic shot noise fluctuations, which mainly affect the pulse phase at low seed power levels ($\sim 10$ kW). However, this is not a concern for the parameters used here, when the electron bunch is driven close to saturation in the first FEL stage (MW seed power; see discussion on self-seeding in SI Appendix). On the other hand, we find rather tight, but nonetheless achievable, tolerances for electron beam parameters, such as the current, energy, energy spread, and transverse offset. All impact the gain length and, thereby, the phase from seeding to saturation. We note that this not only forms a limitation, but also provides a means to tune the phase and amplitude relation of the pulses (Materials and Methods).

**Time-Domain Interferometry**

We turn now to the applications of our proposed X-ray beam splitter. The first is to take advantage of the power spectrum with tunable modulation and phase (Fig. 2C) to characterize absorption lines by their Fourier transforms without moving a monochromator. Spectroscopy can be performed by varying the phase shift and/or the delay between the pulses and collecting integrated counts on a detector (Fig. 4A). This technique is in complete analogy with Fourier transform infrared spectroscopy, where the incident beam is prepared in the spatial, rather than the time, domain by positioning a mirror. Such measurements—now in the X-ray regime—can underpin (optical) pump

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**Fig. 3.** Relative phase stability of the two X-ray pulses as a function of the time delay $\Delta t = t_2 - t_1$, benchmarked by the first-order correlation function $g^{(1)}(t_1, t_2)$. Pink and blue lines show the results assuming a self-seeding chicane with a resolving power of 50,000 and 10,000, respectively.

**Fig. 4.** (A) Schematic of time-domain XRI experiments where an incident pulse pair (green) with a fixed phase relation and time delay $\Delta t$ is transmitted through (pink) or scattered by (purple) a sample. The respective signal is recorded on a two-dimensional (2D) charge-integrating (charge-int.) detector. (B) Frequency-domain RIXS requires high-resolution gratings before and after the sample for incident photon energy selection, analysis of the scattered signal and detection using an array of charge-coupled devices (CCD). Instead, XRI does not require a monochromator after the undulators and can benefit from the multiplexing in momentum transfers of an area detector.
(X-ray absorption)–probe experiments of, e.g., core-hole lifetimes modified via photo-excited modulation of states at the Fermi level. Judicious choices of Fourier components, where \( \Delta E \) is specified by particular values of \( \Delta t, \) allow for highly efficient determinations of key parameters, such as the widths of well-defined lines.

Further possibilities exploiting the modulation illustrated in Fig. 2C and the presence of coherent processes in the sample include resonant scattering performed with and without an analyzer (Fig. 4B). The former enables resonant inelastic X-ray scattering (RIXS), where scans are performed by varying the incident beam modulation for fixed outgoing beam energies, rather than by moving a monochromator, and is complementary to RIXS instrument concepts, where the incident beam energy is encoded spatially on samples (27, 28). Benchmarking of frequency-domain RIXS against time-domain XRI shows that the latter can be a game changer at high-repetition rate X-ray FELs (SI Appendix).

Given that we are working with coherent beams on timescales comparable to core-hole lifetimes, it is interesting and important to consider that there is a similar timescale \( \tau \) for the liberation of electrons from the cores of the atoms used for photodetection. This means, in a semiclassical description, that the detector measures not simply the integral over the square of the impinging electrons from the cores of the atoms used for photodetection. The phase coherence of the pulse pairs guarantees that over the coherence time \( \tau \), the electron from the core is captured by assuming a Heaviside step function.

\[
I = \tau^{-2} \int \int A(t') e^{-(t-t')/\tau} \theta(t-t') dt' \right|_{t}^{2} dt,
\]

where \( \theta(t) \) is a Heaviside step function.

Our “beam splitter” produces optical fields, which can be regarded as sums of delta functions \( \delta(t) \) in time, which can undergo stretching, scattering from or transmission through a sample. Whether or not such stretching occurs, the basic physics of what is measured by the detector is still captured by assuming \( A(t') = A_{1}(t') + A_{2}(t' - \Delta t), \) from which one obtains

\[
I \propto |A_{1}|^{2} + |A_{2}|^{2} + 2 \text{ Re}(A_{1} A_{2}) e^{-\Delta t / \tau}.
\]

The phase coherence of the pulse pairs guarantees that over timescales set by decoherence in the detector, i.e., \( \tau \geq \Delta t \), there will be visible interference terms, which measure directly the correlation between scattering amplitudes at different times for the sample. Thus, if we consider that the momentum transfer \( q \) is defined by the position of the relevant pixel on the detector (Fig. 4A), we are seeing the intermediate scattering functions \( F(q, t) \) of the sample, which is proportional to the Fourier transform in space of the time-dependent, two-particle correlation functions \( G(r, t) \) for generalized charge and/or magnetization densities.

On the other hand, for \( \tau \ll \Delta t \), we will simply see the superposition of the ordinary scattering patterns, i.e., \( I \propto |A_{1}|^{2} + |A_{2}|^{2} \), and we can, by averaging over many pulse pairs, only measure the four-particle correlation function associated with speckle (29). The ergodic theorem tells us that speckle should rigorously vanish in the infinite volume limit for systems at equilibrium (26). However, the requirements for ergodicity are hardly met for many samples of contemporary interest, and our scheme will allow speckle correlations to be measured at unprecedentedly short times. We note, though, that minimal speckle represents an advantage for isolating the interference terms probing the two-particle correlation functions and for following these to \( \Delta t > \tau \).

We turn now to what may well be the most far-reaching implication of phase-locked pairs of subfemtosecond X-ray pulses: the possibility of implementing the full program of quantum optics with X-rays. In particular, our scheme permits tuning of the phase difference and amplitudes of the two pulses (Materials and Methods), enabling coherent control and readout of prepared states, e.g., in photon-echo-type experiments, which at FELs have been demonstrated in the far-infrared (5, 30, 31) and extreme UV regime (8). The latter builds on a combination of phase-modulated seed pulses and high-gain harmonic generation, with beam energy fringing tolerance up to \( E = 47.5 \text{ eV} \). However, this approach cannot delay pulses less than \( \Delta t \approx 150 \text{ fs} \), which is long compared to the decoherence times \( \tau \) for excitations of atomic cores (that is also relevant for the detector, as described above). In contrast, with the PHLUX scheme, pulses are “split” in the electron accelerator, which permits application at higher (soft and hard X-ray) energies and provides access to a larger momentum range. The addition of multiple evenly spaced slits on the microfabricated mask is an extension of the design shown in Fig. 1B and yields trains of phase-stable subfemtosecond pulses—an X-ray frequency comb.

Importantly, using our mode \( \Delta t \) can also be reduced to a few femtoseconds. In combination with the tunability of \( \Delta \phi \), this allows for two few-femtosecond-delayed X-ray pulses with a phase difference of \( \Delta \phi = \pi \). Beyond a critical threshold intensity, self-induced transparency occurs when short coherent light pulses interact with a dense medium, resulting in anomalously low absorption (32–34). Namely, this holds when, within an optical cycle cycle, the total momentum of energy is coherently absorbed by a resonant two-level system, as is coherently emitted thereafter. For the single pulse experiments performed to date, scattering is eliminated along with absorption (35, 36). PHLUX enables generalizations of such experiments to pairs of \( \pi \)-shifted pulses, where the first pulse resonantly excites, and, shortly thereafter, the second pulse resonantly de-excites the sample. Such a sequence could take place on timescales faster than radiation damage and, nonetheless, would allow certain scattered signals to emerge. This would represent another potential route, in addition to ghost imaging (37), to damage-free X-ray scattering, with profound implications for all fields of X-ray science, particularly also for first-principle structure determination of solids and biological samples.

**Materials and Methods**

Simulations of the PHLUX mode were carried out with the three-dimensional, time-dependent FEL code Genesis 1.3 (38), recently released in its fourth version and available at http://genesis.web.psi.ch, using the parameters of the SwissFEL Athos soft X-ray beamline (21) at a seed photon energy of \( E = 1.097 \text{ eV} \). The input was prepared to model the emittance degradation from the mask and the seed signal. This also included variation in the hard X-ray footprint of the two-pulse effects of the PHLUX operation mode less, since \( \Delta \phi \) is given by the monochromator settings (Turning of the Phase Difference and Radiation Power), as well as the electron-bunch phase space portrait going into the undulators after the initial seeding stage.
Also, FEL amplification of the two sliced pulses enhances the amplitude of the radiation by several orders of magnitude, but changes the radiation phase of the seed only very little. Therefore, interference of the two pulses remains constructive at the central wavelength of the seed signal. Only the modulation within the envelope of the power spectrum (Fig. 2. Inset) varies with jitter, but the central line remains unchanged. Moreover, there is also only a weak phase variation if the growth rate of the FEL amplification process between the two slice changes from shot to shot. Since global jitter, such as that of the electron-beam mean energy, affects both slices equally, the phase relation between the two FEL pulses remains unchanged, and, therefore, the stability of the central frequency in the interference spectrum is preserved. Only relative beam parameter jitter has an effect on $\Delta \phi$. Here, the strongest contribution arises from the shot-to-shot fluctuation in the electron-beam parameters due to the micro-bunch instability. In seeded FELs, this becomes apparent in the so-called pedestal of the spectra (18, 19, 43). For our application, this results in a loss of coherence between the two pulses (damping of first-order correlation function $g^{(1)}(t_1, t_2)$ shown in Fig. 3). The micro-bunching instability is difficult to simulate, but its impact on the performance of PHLUX scheme can be estimated from varying the beam parameters. SI Appendix, Fig. S1 shows this dependence of the radiation power and phase relation on the most important sources of jitter. At the SwissFEL Athos beamline, we expect that the relative peak current and energy spread will vary by less than 1% and, thus, do not significantly contribute to jitter. Concerning the relative mean energy and trajectory jitter, the standard requirements to operate self-seeding, e.g., $\langle -10^{-4} \text{ mean energy and } <1 \mu \text{m orbit} \rangle$, stability already imply acceptable phase and power jitter between the two pulses. These estimates are based on the assumption of an optimally configured laser heater (44), which minimizes the impact of the microbunching instability. 

### Tuning of the Phase Difference and Radiation Power

The relative phase difference $\Delta \phi$ between the two X-ray pulses can be controlled by a slight detuning of the self-seeding monochromator: If the beam energy, undulator field, and central wavelength of the self-seeding monochromator are identical, then the phase change along the seed pulse is constant. Detuning of the latter induces a linear spatial change of the seed pulse phase. Consequently, also the sliced portions of the pulse then feature a phase difference given by

$$\Delta \phi = (k_{\text{MC}} - k_{\text{H}})c \Delta t,$$

where $c$ is the speed of light, $k_{\text{MC}}$ and $k_{\text{H}}$ are the central wavenumbers for the self-seeding monochromator and radiator, respectively. For example, a seed photon energy of $1.1957 \text{ eV}$ and time delay of $\Delta t = 7 \text{ fs}$ requires a relative detuning of $2.7 \times 10^{-4}$ for a pulse-to-pulse phase difference $\Delta \phi = \pi$. Such manipulation is difficult for pulse pairs that overlap in time, but else this variant of the PHLUX scheme does not represent a major restriction in terms of operation. The total radiation power tunable by use of a so-called laser heater (44) or by removing/adding undulator modules that contribute to the lasing process. The relative amplitude or rather radiation power $\Delta P$ of the two pulses can then be varied by, for example, adding a transverse or energy chirp to the beam. That way, the radiation power of the two phase-locked pulses can be individually adjusted, e.g., for Ramsey ($\pi/2 - \pi/2$) or Hahn-echo ($\pi - \pi$) type experiments.

### Data Availability

All study data are included in the article and/or supporting information. 

Note Added in Proof. In the course of the review process, we became aware of the recent demonstration of phase-stable hard X-ray pulse pairs based on superluminescence and self-seeded free-electron laser, which underlines the importance of the more general working principle, and enabled experiments reported thereon (46).

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