ENGINEERING ADVANCEMENTS IN X-RAY PUMP–PROBE TECHNIQUES: DELAYLINE FOR ATTOSECOND SCIENCES AT LCLS

Hengzi Wang*, Taran Driver, Yoshio Ichii, Namrata Balakrishnan, Randy Whitney,
James Cryan, Agostino Marinelli
SLAC National Accelerator Laboratory, Menlo Park, CA, USA

Abstract

X-ray pump–probe experiments at X-ray free-electron laser (XFEL) facilities, such as LCLS, enable investigation of ultrafast electronic and atomic dynamics. Central to these experiments are advanced delayline systems for generating X-ray pulse pairs with finely tunable time delays and precise alignment. In this work, we present background on delayline techniques and focus on the development of the next-generation Soft X-ray (SXR) delayline at LCLS. The SXR delayline delivers attosecond- to femtosecond-resolution timing, exceptional beam stability, and rapid mode switching, addressing the limitations of earlier technologies. We discuss its optical engineering challenges, control strategies, and scientific applications in ultrafast materials, chemistry, and quantum science.

INTRODUCTION

Ultrafast X-ray science, enabled by X-ray free-electron lasers (XFELs) such as the Linac Coherent Light Source (LCLS), has revolutionized our ability to observe atomic, molecular, and condensed matter dynamics at unprecedented time scales, extending into the attosecond regime [1]. Central to these advances are X-ray pump–probe experiments, which rely on precise control and generation of two time-delayed X-ray pulses to explore ultrafast phenomena that were previously inaccessible.

To achieve this precision, various delayline systems have been developed at LCLS, utilizing both magnetic chicane architectures and split-and-delay optics. Magnetic chicanes employ sequences of dipole magnets to introduce variable path length separations in the electron beam, thereby generating two X-ray pulses with adjustable temporal delays [2]. Split-and-delay optics utilize high-quality crystals and mirrors to split a single X-ray pulse into two, directing each along distinct optical paths with engineered delays before recombination at the sample [3]. Both technologies present important trade-offs in terms of achievable delay range, temporal resolution, and beam stability. Typically, these techniques can deliver delays from several femtoseconds to the nanosecond scale.

However, for attosecond science, new approaches are required. At LCLS, the XLEAP scheme was developed to deliver attosecond X-ray pulses; nevertheless, the pulse duration in XLEAP is not tunable, and the scheme lacks the ability to freely adjust delays across zero or to independently shape or steer both X-ray pulses. These limitations constrain its suitability for next-generation attosecond pump–probe experiments [4].

To address these needs, the Soft X-ray (SXR) delayline project was launched at LCLS, targeting the soft X-ray regime with tunable sub-femtosecond delays, robust operational stability, and rapid mode-switching capabilities. The SXR delay line is installed in the SXRSS section between the 3rd and 4th dipole magnets (see Fig. 1), providing tunable time separation for the generation of X-ray pulse pairs. Its optical path comprises a pair of ultra-precision mirrors mounted on translation stages, actively aligned via piezo actuators and monitored with two capacitive sensors, achieving attosecond timing fidelity.

This integrated system overcomes the limitations of both XLEAP and SXRSS, delivering attosecond-resolution delay tuning, high repeatability, and long-term stability. As a result, it enables new advances in attosecond-resolved studies of quantum materials, ultrafast chemical reactions, and nonlinear X-ray interactions.

SXR DELAYLINE: MOTIVATION AND SCIENCE DRIVERS

The SXR delayline is conceived to overcome key shortcomings and unlock new science opportunities:

- **Freely tunable delays crossing zero:** Crucial for time-resolved studies where both pump-before-probe and probe-before-pump processes are relevant.
- **Attosecond to few-femtosecond resolution:** Required to follow electron dynamics and transient structure formation.
- **Decoupled pulse properties:** Ability to independently specify the energy, arrival time, direction, and duration for both X-ray pulses, broadening the experimental toolkit.
- **Long-scale stability and repeatability:** Vital for extended beamlines (up to 100 m), where micron-scale misalignments can destroy time resolution and focal overlap.
- **Rapid mode switching:** Support for soft x-ray self-seeding (SXRSS), SXR delayline pump–probe, or normal SASE[1] operation.

---

* hengziwang@slac.stanford.edu

[1] SASE: Self-Amplified Spontaneous Emission Free-Electron Laser. SASE FELs generate intense coherent X-ray pulses via exponential amplification of spontaneous emission in an undulator, as the electron bunch propagates without external seeding.

The instrument is designed to support a broad range of ultrafast science, from electron transfer in molecules to phase transitions in quantum materials and nonlinear X-ray photon interactions.

ENGINEERING DESIGN AND ARCHITECTURE

The SXR delayline integrates mechanical, optical, and electronic control innovations to meet its demanding specifications.

Mechanical and Optical System

The delayline is located downstream of a four-dipole-magnet chicane (Fig. 1). The chicane introduces a variable path difference for the electron beam, providing a tunable interval between X-ray pulses generated in separate undulator sections.

A set of ultra-precision mirrors form the core of the delayline optical path. Mirrors are fabricated with surface figure errors less than 0.15 nm RMS and slope error less than 0.1 μrad RMS and surface roughness under 0.1 nm RMS. To preserve attosecond time resolution and wavefront quality, the mirrors are mounted on translation stages capable of sub-nanometer positioning. For a 1 nm linear shift, the timing difference imparted on the pulse is approximately 3 attoseconds.

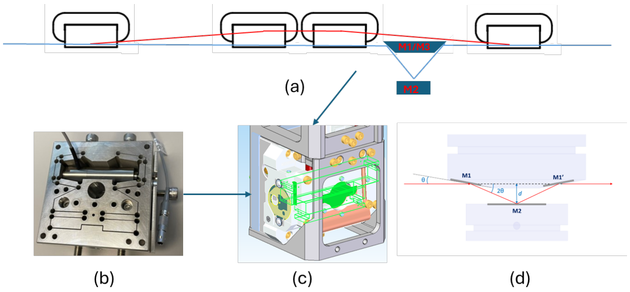


Figure 1: Schematic of the SXR delayline. (a) SXRSS section: the electron beam (red) is bent by a four-dipole-magnet chicane, generating two X-ray pulses with variable delay; (b) mirror mover powered by piezo actuator; (c) precision aligned mirror pair, one convex and one flat; (d) the two mirrors form a three-bounce path that generates delays.

The delay time (t) can be calculated as follows:

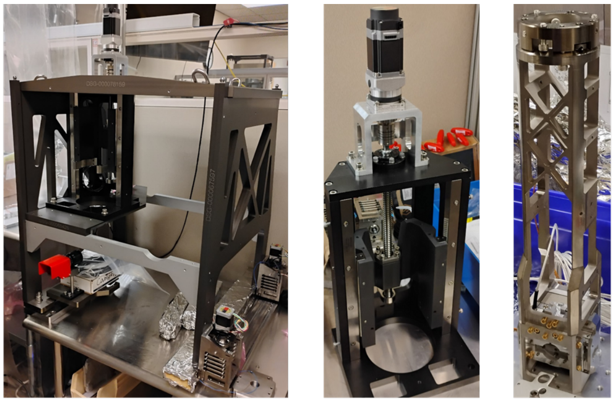
$$t = \frac{2d(1 - \cos(2\theta))}{c \cdot \sin(2\theta)} = \frac{2d \cdot \tan(\theta)}{c} \tag{1}$$

where θ is the mirror incident angle and c is the speed of light.

Active Alignment and Diagnostics

Achieving and maintaining precise alignment is critical for attosecond experiments, where microradian misalignments can severely degrade performance. Conventional mechanical alignment is insufficient for this level of sensitivity. To address this, the mirror assembly, together with the vacuum chamber, can be driven by two linear actuators positioned at the far end of the support frame, enabling real-time adjustment with sub-microradian resolution. A pair of capacitive sensors continuously monitors both the position and angular orientation of the mirror pair, providing active feedback for compensation of vibration and thermal drift (Fig. 2).

Alignment routines combine mechanical pre-adjustment with beam-based optimization, utilizing downstream diagnostic signals to fine-tune the optical path and ensure stable overlap at the sample position.



Support frame and Pitch correction Linear translation Optical cage

Figure 2: Detailed view of the delayline mirror assembly. Dual piezo actuators (red) control beam-based alignment; capacitive sensors (blue) continually monitor position and angular drift. The modular optical cage supports sub-microradian and nanometer-level control for attosecond timing precision, facilitating automatic switching between delayline and self-seeding configurations.

Optical Cage and Flexible Operation Modes

A distinctive feature is the shared optical chamber with the SXRSS slits. Linear stage mechanisms enable rapid switching between three configurations (Fig. 3):

- Self-seeding with SXRSS slits;
- Delayline pump–probe mode;
- Beam-stay-clear mode for calibration or alternate optics.

This modularity ensures user access and experimental agility, a significant improvement over previous approaches.

SYSTEM IMPLEMENTATION AND PERFORMANCE

The SXR delayline at LCLS is currently under construction, with initial testing focused on the piezo actuator motion system. Early results indicate that the actuator assembly can reliably achieve position adjustments with a resolution on the order of a few nanometers.

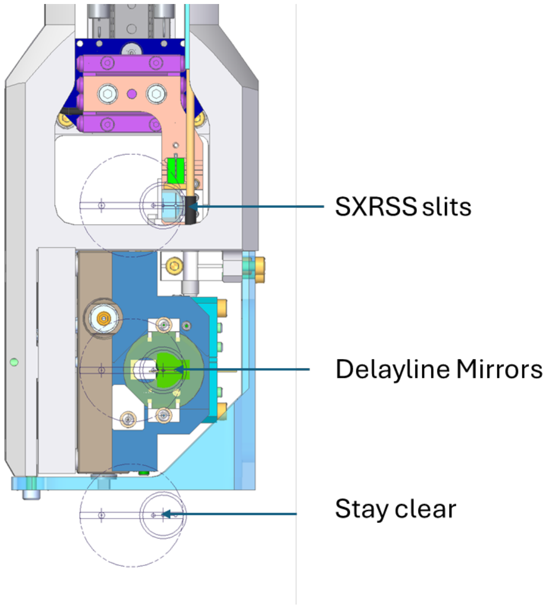


Figure 3: Driven by a linear actuator, the system can quickly switch between SXRSS, SXR delayline, and normal SASE operation.

- **Delay range:** The final system is designed to enable continuously tunable delays in attosecond steps, with a targeted range up to ±5 fs (goal: ±8 fs).
- **Resolution:** Initial tests of the piezo actuators demonstrate stable and repeatable nanometer-scale positioning.
- **Spatial alignment, stability, and automation:** These aspects are planned for future implementation and testing; corresponding diagnostics, feedback systems, and automated routines are under development.

Further commissioning and system integration are in progress, with expanded performance characterization planned for subsequent experimental phases.

CONCLUSION

The SXR delayline at LCLS exemplifies a new generation of XFEL instrumentation: tunable, precise, and robust control of X-ray pump–probe delays at attosecond scales, with automated alignment and switchable operation modes. These advances unlock transformative experiments spanning quantum physics, femtochemistry, and materials science, and serve as a technical foundation for future upgrades and emerging scientific frontiers.

ACKNOWLEDGEMENTS

We gratefully acknowledge the contributions of the LCLS engineering, optics, and EED controls teams. Funding for this research is provided by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences under Contract No. DE-AC02-76SF00515.

REFERENCES

[1] P. Emma *et al.*, “First lasing and operation of an angstrom-wavelength free-electron laser”, *Nat. Photonics*, vol. 4, no. 9, pp. 641–647, 2010. `doi:10.1038/nphoton.2010.176`

[2] D. Ratner *et al.*, “Experimental demonstration of a soft x-ray self-seeded free-electron laser”, *Phys. Rev. Lett.*, vol. 114, no. 5, 2015. `doi:10.1103/PhysRevLett.114.054801`

[3] D. Zhu *et al.*, “Development of a hard x-ray split-delay system at the linac coherent light source”, in *Proc. SPIE’17, 10237 Advances in X-ray Free-Electron Lasers Instrumentation IV*, Prague, Czech Republic, pp. 1–8, 2017. `doi:10.1117/12.2265171`

[4] J. Duris *et al.*, “Tunable isolated attosecond x-ray pulses with gigawatt peak power from a free-electron laser”, *Nat. Photonics*, vol. 14, no. 1, pp. 30–36, 2020. `doi:10.1038/s41566-019-0549-5`