Low-loss Stable Storage of X-ray Free Electron Laser Pulses in a 14 m Rectangular Bragg Cavity

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Abstract

We present an experimental demonstration of a stable, low-loss, large x-ray cavity operating at 1.2 Angstrom wavelength. The cavity consists of 4 high reflectivity single crystal diamond Bragg mirrors arranged in a rectangular configuration with a design round trip distance of 14.2 m. Femtosecond x-ray laser pulses from an x-ray free electron laser were coupled into the cavity via a transmission phase grating. The cavity round trip efficiency and lifetime were characterized quantitatively, indicating close to the theoretical optical performance. By introducing focusing optics, we show that a stable cavity mode can be trapped within. The direct observation of sustained stable x-ray circulation provides the most direct evidence to date that cavity-based x-ray free electron lasers and other cavity-based hard x-ray systems are feasible.

One-Sentence Summary

We report on first experimental demonstration of a large-scale x-ray laser cavity operating at 1.2 Angstrom wavelength.

Main Text

In 1958 Schawlow and Townes proposed an optical maser based on a gain medium introduced in a cavity with a single resonant mode (1). Their work paved the way for modern laser oscillators, which have led to countless applications in today's science and technology. Optical cavities, near ubiquitous amongst laser systems, typically play a large role in defining a laser's properties, including its exquisite degree of temporal coherence. Unsurprisingly, the first free-electron lasers (FELs) were low-gain oscillators, or FELOs, that utilized a cavity and operated at infrared, visible, and ultraviolet wavelengths (2,3,4). While the oscillator concept was considered the most straightforward to scale towards shorter wavelengths (5,6) when x-ray FELs were first conceived, the technology for constructing a large cavity at Angstrom wavelength was considered out of reach. Therefore, all current x-ray FELs (XFELs) have adopted the single pass, high gain design. This results in relatively poor temporal and spectral coherence of the emitted field, owing to the fundamental stochastic nature of the electron bunch dynamics. Hard x-ray self-seeding has been applied to improve coherence but still suffers pulse energy instability (7–12). Hard x-ray pulses with the full spatial temporal coherence and high stability demanded by many novel x-ray laser experimental techniques have yet to be demonstrated at any XFEL facility.

Many cavity-based conceptual design proposals for FELs have been studied, all projecting many orders of magnitude improvement in brightness and stability. In fact, the first proposal to use Bragg reflectors to circulate x-ray radiation was made in 1984 (12), about the same time the single-pass high-gain FEL concept was proposed (14). Huang and Ruth proposed in 2006 to use a high-gain FEL in a Bragg cavity to form an x-ray regenerative amplifier XFEL (XRAFEL) (15,16,17). Kim et al. proposed in 2008 to use a low-gain FEL to construct an oscillator (XFELO) (18). More recently, Halavanau et al. proposed to use the Bragg cavity for a population-inversion x-ray laser oscillator (XLO) (19). Upcoming experiments at both
LCLS and EuXFEL will provide early tests of these concepts (20, 21). In almost all proposals, Bragg optics, in particular single crystal diamonds, have been adopted as the cavity optics due to their potential high reflectivity, and superior thermal mechanical properties. This has since been experimentally verified via technological breakthroughs in the growth of defect and strain-free synthetic diamond crystals (22,23). A natural next step is to experimentally verify the performance of a Bragg x-ray cavity suitable for a cavity-based XFEL, which can not only select a single longitudinal mode for amplification but also provide substantial suppression for undesired modes.

In contrast to the typical tabletop scale cavities of optical lasers, x-ray cavities for a cavity-based XFEL (CBXFEL) will require a much larger physical dimension, often approaching hundreds of meters in length. This footprint is driven by the physics of the gain media and gain process of an XFEL. Amplification requires interaction between x-ray pulses with a train of relativistic electron bunches through long undulators. The repetition rate of the electron source, dictating the cavity round trip time, is governed by available high brightness electron accelerators. For a modern superconducting linear accelerator with a MHz scale repetition rate, this sets the cavity length on the order of 100 m. The large cavity dimensions, in turn, demand very tight spatial and angular tolerances and requirements on the Bragg mirror system.

Here we report the performance of a 14.2-m round-trip-length rectangular Bragg cavity operating at 1.261 Angstrom as a critical demonstration towards realizing a full-scale CBXFEL. We show excellent short and long-term cavity alignment and stability by the observation of nearly 60 single pulse x-ray roundtrips over a 2.8 µs time span, with minimal performance degradation over a period of 1 hour. We investigate transverse cavity dynamics and the effects of intra-cavity focusing and show that cavity losses are close to theoretical values. The performance achieved by this x-ray cavity prototype offers the most definitive demonstration to date that the realization of large x-ray cavities and CBXFELs is very much feasible.

The 14.2-meter-round trip x-ray cavity was constructed and tested at the X-ray Pump-Probe (XPP) instrument at the Linac Coherent Light Source (LCLS) (24). The operation wavelength was chosen at 1.261 Angstrom (photon energy 9.831 keV) to enable high-efficiency Bragg reflection of diamond (400) at 45.000 degrees. The XPP instrument monochromatized the input x-ray beam to a 0.6 eV bandwidth (25), and the beam divergence is approximately 3 µrad FWHM when wavefront distortion from the monochromator is considered in addition to the natural FEL divergence (26). Figure 1 shows the overall optical layout of the cavity. The X-ray beam enters the cavity by first transmitting through the diamond crystal C\textsuperscript{*4} and then through a transmission phase grating with 1 µm period and 1.4% first order diffraction efficiency (27). The +1st diffraction order, which is deflected 126 µrad by the grating, is chosen as the in-coupled beam. It is reflected by the first Bragg mirror (C\textsuperscript{*1}) at exactly 45.000-degree incidence angle. Other grating diffraction orders are outside the angular acceptance window of C\textsuperscript{*1} and therefore transmit downstream. The reflected +1st order, with its bandwidth reduced to ~ 100 meV by the spectral acceptance of C\textsuperscript{*1}, then reflects sequentially through C\textsuperscript{*2}, C\textsuperscript{*3}, and C\textsuperscript{*4} along the rectangular trajectory and arrives at the transmission grating again. This time, the directly transmitted 0th order, with 93.4% efficiency (considering both absorption loss and diffraction loss) through the grating is chosen
and carefully aligned to be co-linear with the $+1$st order diffraction from the incoming beam. This enables the beam to be reflected by $C^*_1$ again and subsequently recirculate in the cavity. A beryllium refractive lens \((28)\) with a 71 m focal length is inserted to further stabilize optical modes in the cavity. The recirculated beam path is enclosed by two vacuum chambers connected with beam transport tubes to eliminate air absorption losses. The 1st order diffraction from the first roundtrip return beam as well as the later roundtrips are separated in space and angle from the on-axis cavity recirculating beam after \(\sim 7\) m of propagation distance. Other unwanted diffraction orders are filtered out with x-ray slits before a selected beam sample exits the cavity vacuum system through a Kapton window. An x-ray profile monitor with nanosecond gating is used to record the average beam position and profile after each round trip, while simultaneously, the x-rays transmitting through the thin scintillator screen are collected by a fast photodiode that records each sample “train” of the recirculating x-ray pulses. This allows for detailed dynamical measurements of the stored x-ray beam pulse property evolution over many passes.

Details of the cavity-loss mechanisms are critical in determining both the lasing threshold and the lasing performance characteristics in all CBXFELs. The overall roundtrip loss, in combination with XFEL gain, determines how many round trips the system requires to reach saturation, as well as saturated output level. We evaluate the observed cavity round trip losses by fitting a waveform to each peak in the averaged photodiode traces and comparing the amplitudes of all neighboring peaks. Figure 2 (A, B) shows these measurements after cavity alignment is optimized. In early passes, the trapped radiation undergoes spectral-angular filtering from the Bragg reflection process and incurs higher losses due to deviation from ideal alignment and cavity mode matching. We observe the per-round trip efficiency grow from 10% quickly to above 80%, as the fraction of matched cavity mode dominates the recirculating photon flux. This process reaches a quasi-steady state with roundtrip efficiency asymptotically approaching \(\sim 88\%\). In this regime, considering the measured 6.6% loss from the in/out coupling grating for the 0th order, and an estimate of 2% absorption loss from the lens, the matched mode has a round trip efficiency > 96%. This indicates reflectivity for each individual diamond Bragg reflection of > 99%, matching theoretical calculation from dynamical diffraction theory. The shaded area in Fig. 2B are enclosed by numerical beam propagation modeling results (see supplemental material) with the upper boundary representing the case of ideal initial alignment and the lower boundary representing a compounded 4 µrad misalignment of the in-coupled x-ray beam from the cavity optical axis in the dispersion plane. The near-perfect round trip efficiency observed at later round trips indicates that the quality of the synthetic high-pressure-high-temperature (HPHT) type II-a diamonds (Sumitomo Electric) is of sufficient quality serve as Bragg mirrors for cavity-based XFELs. While the initial alignment error can lead to longer build up time for an x-ray laser cavity, as long as losses are overcome by the XFEL gain process, eventually the matched cavity mode will be amplified and dominate via optical gain guiding mechanisms \((16, 29)\). The saturated XFEL performance after initial build-up will therefore be less impacted by the early roundtrip loss, but largely determined by the observed later roundtrip efficiency.

The pulse energies at later round trips are extremely sensitive to the cavity alignment, serving as a direct indicator of the cavity’s long-term stability. We found the cavity was able to maintain the average
recirculating pulse energy at the 25-th and the 40-th round trip to within 20% of the initial value as shown in Fig. 2C. When compared to the numerical beam propagation model, this indicates overall angular alignment stability to < 400 nrad over the course of the 1-hour measurement.

The ns-gated microscope provides direct measurement of the average beam position and size information after each round trip. The extracted beam profile images for sequential round trips are shown in Fig. 3A and B for two different cavity configurations: A only using the 4 Bragg mirrors and B with an f = 71 m focusing lens inserted. We find that in the case without intra-cavity focusing, while the beam position was optimized to relatively stable in both horizontal and vertical direction (Fig. 3C and D), the vertical beam size quickly diverged in vertical (Fig. 3F). The increase in vertical beam size is faster than the natural divergence of the incoming FEL beam, indicating a compounded equivalent defocusing effect from the beamline monochromator and cavity diamond optics. In the horizontal direction, the beam size decreases from the ~ 350 µm initial value down to a size of 200 µm, a result of slight focusing effect from the cavity crystals interacting with angular filtering of the Bragg reflections. When the intra cavity focusing lens is inserted, a stable cavity is formed. We observe distinct beam size and position modulation in the vertical direction. Both beam size and trajectory oscillations in the non-dispersive (vertical) plane can be modeled with ray optics (Fig. 3E and F) indicating effective intracavity focusing with f = 93 m. The observed beam size oscillations are analogous to circular particle accelerators (sometimes referred to as betatron oscillations in accelerator physics) (30,31,32). In the dispersion plane, due to dynamical diffraction, we again observed the effects of spectral-angular filtering, with the cavity mode size quickly reaching quasi-steady-state parameters. We note that in CBXFELs, electron beam transverse size and divergence can be adjusted to optimize the roundtrip performance and alleviate the negative effects of the spectral-angular mismatch, by fine-tuning the magnetic transport lattice of the undulator.

In summary, we have experimentally demonstrated for the first time an operating large scale x-ray cavity, based on four Bragg reflections, capable of storing x-rays for up to 2.8 µs with high efficiency that can be maintained for long periods of time. This set the foundation for more sophisticated cavity designs, such as bow-tie cavities (18), or cavities with gain, e.g., cavity-based XFEL, XFELo, or XLO. Intra-cavity focusing was shown to be effective in both selecting a well-defined recirculating mode and maintaining the cavity stability. An intra-cavity transmission grating was shown as a simple and efficient way to in-couple and out-couple cavity radiation. Other applications of this setup include benchmarking of low-loss x-ray cavities for future light sources based on storage rings with cavity-based FEL oscillators (33), gamma-gamma scattering (34) imaging and quantum optics experiments (34), where high coherence and high-repetition-rate is required.

**Materials And Methods**

**Cavity Optics**
Cavity Bragg mirrors were HPHT type II-a single crystal diamond plates of thickness ranging between 270 to 600 µm, with C\textsuperscript{*} being the thinnest. All crystals surfaces are cut and polished along the (100) crystal planes, with strain-relief cuts added by laser micro machining in order to reduce strain from clamping (36). The diamonds were characterized and selected based on rocking-curve imaging and surface asymmetry measurements to ensure a greater than 1 × 1 mm area with < 0.2 µrad strain and < 0.5° miscut. The crystal characterization process is described in more detail in (37, 38). The diamond transmission grating was fabricated on a CVD polycrystalline diamond substrate with a thickness of 30 µm using reactive ion etching. The grating period was 1 µm with an optical window size of 1 mm by 1 mm. The etch depth was chosen targeting a 1–2% first order diffraction efficiency at 9.831 keV.

**Cavity Diagnostics**

Initial crystal Bragg condition was established using large area Canberra PIPS diodes positioned in the reflected beam path for each crystal. Time resolved ring-down trace was measured using a high speed photodiode (Optodiode AXUV20HS1) with rise/fall time well below 10 ns when reverse biased. The signal is amplified using a broadband RF amplifier and recorded at 2 GSps by an 8-bit digitizer (Agilent Acqiris DC282). An Andor iStar sCMOS camera with UV imaging optics was used to spatially and temporally resolve the x-ray beam profile after each roundtrip. Single crystal Ce:YAP scintillator of 30 µm thickness was used, taking advantage of its short fluorescence lifetime of ~ 25 ns at 370 nm. The combined time resolution of the imaging system approximately 32 ns as shown in Fig. S2. The sampled x-ray flux ($10^4$ photons/pulse) for later round trips is too low to allow single-shot imaging measurements. Multi-pulse exposure with synchronized electronic gating was used to obtain beam profile measurements shown in Fig. 3. Fluorescence background subtraction from earlier roundtrips was removed according to process shown in Fig. S3.

**Cavity Alignment Procedure**

The initial diamond crystal orientation alignment was sequentially established by inserting large-area photodiodes after each crystal and maximizing the reflection. Once the first-round-trip return was established, the diamond mirror Bragg angles were adjusted to guide the beam towards the 0th round-trip beam position using the ns-gated camera by iteratively steer the diamond Bragg mirrors. The exact photon energy is initially chosen by adjusting the upstream monochromator and changing the angles of $C_{1,2}^\ast$, to make the long sides of the rectangular beam path equal distance in the horizontal plane at the two ends of the cavity. Once the correct photon energy was identified, waveform from the fast diode revealed the first few roundtrips. The final iterations involved micro adjustments of position and angle of diamond Bragg mirrors to maximize the number of roundtrips seen in the ringdown waveform, and the pulse intensity of the last-round-trips. After optimal alignment was achieved, beryllium lens ($f = 71$ m) was inserted to stabilize the beam trajectory inside the cavity. The angular alignment precision provided by the motorized stages were ~ 500 nrad in the dispersion plane (horizontal) and ~ 50 nrad in vertical. Each crystal alignment based on rocking-curve scan carries ~ 1 µrad uncertainty, contributing to lower pass-by-pass efficiency in the early round trips. However, it does not impact the steady state cavity efficiency for the “resonant” cavity mode as seen for later roundtrips. See Fig. 2B.
Declarations

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Competing interests: The authors declare that they have no competing interests.

Data and materials availability: The data that support the findings of this study are available from the corresponding author upon reasonable request.

References And Notes


**Figures**

**Figure 1**

*Experimental schematic of the cavity optics and diagnostics.* Four diamond Bragg mirrors are used as the main cavity optics. A portion of the incoming x-ray laser beam (purple) is injected into the cavity by a diamond transmission grating using its 1st order diffraction (red). The red rectangle indicates the beam trajectory inside the cavity once the correct photon energy was chosen and the Bragg mirrors are aligned. A beryllium lens can be inserted into the beam path to stabilize the beam propagation over many round
trips. The orange beam path indicates the 1\textsuperscript{st} order diffraction of the circulating beam as it passes through the diamond grating each time. This beam was isolated and used for beam dynamics monitoring with a high-speed imager consisting of a fast scintillator and a nanosecond-gated microscope. The transmitted beam through the thin scintillator is measured by a downstream photodiode.

**Figure 2**

**Cavity ring-down measurement.** (A) Averaged ring-down measurements of the cavity with intra-cavity focusing ($f=71\text{m}$). 59 total roundtrips are shown within the 2.8 $\mu$s measurement window. The inset shows an estimate of the average number of photons remaining in the cavity after each round trip. (B) Cavity round trip efficiency. The shaded area between the dashed lines is an estimate from a numerical model assuming perfect alignment and alignment with a 4$\mu$rad initial angular error. (C) Normalized average pulse intensity after round trip 25 and 40 over a one-hour period.
Figure 3

Transverse beam circulation dynamics. (A and B) Beam spatial profile evolution in the x-ray cavity for with and without inserting intra-cavity focusing lens. The scale bar indicates 200 μm. (C and D) Beam transverse position after each roundtrip in horizontal and vertical directions. (E and F) Beam transverse size (full width) after each round trip in horizontal and vertical directions.

Supplementary Files

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