5-4. SACLA Beamlines

1. Operation status

Recovering from the significant impact of the COVID-19 pandemic in FY2020, the FY2022 operations of all three beamlines (BL1–3) of SACLA have recovered to close to those before the pandemic. The user beamtime had almost recovered in FY2021 and remained at a similar level (~6,000 hours) in this fiscal year. Note that one of the three beamlines, BL1, is a soft X-ray (SX) free-electron laser (FEL) beamline using a dedicated linac operated at 60 Hz. The other two beamlines, BL2 and BL3, are hard X-ray FEL (XFEL) beamlines sharing the main linac. The two XFEL beamlines are operated simultaneously by using a fast-switching magnet that changes the electron beam route in a pulse-by-pulse manner. Each XFEL beamline provides X-ray pulses with a 30 Hz repetition rate when the accelerator is operated at the maximum rate of 60 Hz. The switching magnet also delivers the electron beam to the storage ring of SPring-8.

The major topics regarding upgrades on beamlines and experimental stations in this fiscal year are summarized in the following subsections, including some achievements resulting from close collaborations with external experts through programs such as the following.

- SACLA/SPring-8 Basic Development Program
- SACLA Industry–Academy Partnership Program
- SACLA Research Support Program for Graduate Students

2. SX-FEL beamline (BL1)

In FY2022, we have been developing a flat-field grazing-incidence spectrometer and nanofocusing systems at the SX FEL beamline (BL1) under the support of the Basic Development Program.

The grazing-incidence spectrometer is equipped with an aberration-corrected concave grating with a groove density of 1200 lines/mm and a thermoelectrically cooled CCD camera. The spectral resolution (E/ΔE) is ~1800. The CCD camera captures data with a collection rate of 60 Hz under a full-vertical binning mode. The data are stored in the SACLA HPC storage system, which allows easy access to the data and integration with various types of SACLA data such as timing monitor data. The spectroscopy is feasible for the fundamental FEL radiation and also the 3rd- or 5th-order harmonics [1].

Two types of nanofocusing system are under

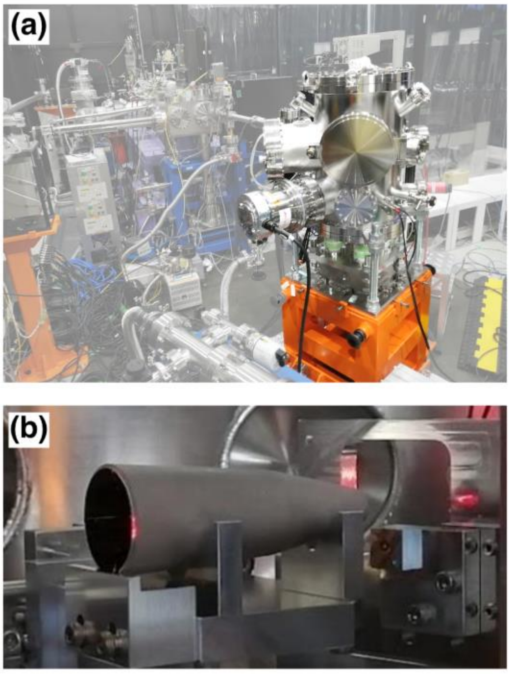

Fig. 1. (a) Photograph of the general-purpose nanofocusing apparatus equipped with (b) a focusing Wolter mirror.

development at BL1. One is a general-purpose nanofocusing system (Fig. 1) capable of stably forming a beam with a diameter of around 400 nm. The other is a wavelength-scale focusing device targeting a beam diameter of 50 nm. The general-purpose nanofocusing system has been preliminarily utilized in user experiments, yielding results in nonlinear signal measurements in the soft X-ray region. In the commissioning of the wavelength-scale focusing device, we have not yet reached the diffraction-limited focusing size. However, we have carried out proof-of-concept experiments using the unique light propagation properties of a hollow beam for a weak-signal detection scheme. We will continue to develop the fabrication apparatus to improve the accuracy of the surface figure of the focusing mirrors. We aim to launch this development within FY2023 and hold high expectations for realizing higher precision mirrors and more advanced focusing devices.

3. XFEL beamlines (BL2 and BL3)

3-1. Nanofocusing optics

The user operation of the sub-10 nm focusing system based on Wolter III optics started in FY2022. Developed in collaboration with Prof. Kazuto Yamauchi's group at Osaka University, this system is aimed at generating an X-ray laser field with an unprecedented intensity of $\sim 10^{22}$ W/cm^2. The precise alignment of its focusing mirrors is achieved and maintained using wavefront sensing techniques. This system is designed for a photon energy of 9 keV and is available in BL3 EH4c. User experiments in X-ray nonlinear optics and high-energy-density science have been steadily performed in this fiscal year.

Apart from this most advanced tight focusing system, a portable compact nanofocusing system based on Kirkpatrick–Baez optics has been developed. In this compact system, the total length from the upstream edge of the first elliptical mirror to the focal point is only 340 mm. During the performance test experiment, focusing on 240 nm and 360 nm for the horizontal and vertical directions was achieved. Further reduction of the focal spot size is expected by employing more precise stages in the system alignment. After the commissioning of the focusing system with the improved stages in 2023, the system will open for user experiments at either BL2 or BL3. The compact focusing system is expected to be utilized for not only tight-focused X-ray experiments but also optical pump and tight-focused X-ray probe experiments.

3-2. Accessibility Improvements of synchronized femtosecond laser systems

Pulse lasers with a temporal duration of a few tens of femtoseconds are extensively used in ultrafast pump and probe experiments at XFEL facilities. Two sets of millijoule-class Ti:sapphire laser systems located in LH1 have been utilized at SACLA. The stable operation of these laser systems is critical for the success of a broad range of research activities.

The two laser systems are often operated in parallel for independent experiments, one at BL1 and the other at BL3. Therefore, the occasion of accessing LH1 is severely limited to avoid any influence on the laser stability, such as room-temperature fluctuations. Laser maintenance and tunings are performed mostly on Mondays when no user experiment is scheduled. Because of the large demand for the use of the femtosecond lasers, it is sometimes difficult to reserve sufficient maintenance and optimization time. Furthermore, the current situation of having two laser systems in a laser hutch may cause significant difficulties when a system requires urgent long-term tuning work.

To manage the difficulties, a new laser hutch (LH2) has been constructed to house the laser systems in individual hutches. The laser system delivering the laser to BL1 will be relocated to LH2 in the summer of 2023. The laser is expected to resume its user operation immediately in the latter half of FY2023.

3-3. Extension of experimental capabilities at the laser compression platform

The front end of a high-power nanosecond laser system has been upgraded to stabilize the output energy and extend the capability of generating arbitrary waveforms. The stabilized shot-to-shot jitter of the pulse energy and power is 1.3% and 3% root-mean-squared (rms), respectively. This output stabilization allows us to proceed with more precise arbitrary waveform generation. The upgraded front end is now capable of generating pulses with a width of up to 20 ns. The upgraded system will be available for users in FY2023. The facility plans to offer users a simple temporal waveform initially and aims to develop more complex waveforms to explore a broader range of pumped states.

In order to expand the coverage of X-ray diffraction measurements at the laser compression platform, the facility has installed an additional detector in the experimental chamber. This system is anticipated to be ready for use in FY2023.

3-4. Advances in hard X-ray split-and-delay system

A hard X-ray split-and-delay optical (SDO) system consisting of four Si(220) Bragg crystals has been developed at BL3, enabling advanced X-ray measurement and diagnostic techniques using monochromatic XFEL pulse pairs with a tunable and jitter-free time separation of up to ~200 ps. Thanks to the self-seeding mode of operation [2] and the standard 1 μm focusing system at EH4c of BL3 [3], the SDO system has achieved a high intensity of ~10^{17} W/cm^2 (~10 μJ/pulse) with a relative pointing jitter of ~0.1 μm rms on samples. The high intensity and stability allowed for the completion of second-order autocorrelation measurements, which can be used to evaluate the pulse duration of the monochromatic XFEL pulses, directly. The measurement with two-photon absorption in a zirconium thin film indicated that the pulse duration was ~7.5 fs FWHM at 9 keV [4]. Some unique experiments, such as picosecond-scale XFEL pump–XFEL probe studies, were also successfully performed using the SDO system in FY2022.

3-5. Improvements of the experimental instrument for nanobeam coherent diffractive imaging

An advanced multiple application X-ray imaging chamber (MAXIC-S) has been developed for a coherent diffraction imaging (CDI) experiment using a 100 nm XFEL beam produced with a multilayer-mirror focusing system. It was demonstrated that 4 keV XFEL was focused down to 60 nm × 110 nm and a 2 nm resolution was achieved in CDI of metallic nanoparticles [5]. The MAXIC-S system has been upgraded for more reliable and user-friendly operation. For example, the load-lock sample transfer system was motorized and the sample-scanning system was improved for more efficient measurement.

4. Research highlights

4-1. Structural characterization of dynamic control of sequential electron and proton transfer events in a flavoenzyme

Photoreduction of DNA photolyases, utilizing blue light, involves two single-electron transfers to the flavin adenine dinucleotide (FAD) with an intermittent protonation step to prime the enzyme active for DNA repair. Maestre-Reyna et al. described the structural characterization of the FAD post–electron transfer (ET) in the *Methanosarcina mazei* class II DNA photolyase (MmCPDII) via nanosecond-to-microsecond-range time-resolved serial femtosecond crystallography [6]. The obtained molecular videos demonstrate how the protein environment of redox cofactors organizes multiple electron/proton transfer events in an ordered fashion, which could be applicable to other redox systems, such as photosynthesis.

4-2. Generation of phase-stable femtosecond X-ray pulse pairs through superfluorescence

The generation of phase-stable femtosecond X-ray pulse pairs will advance nonlinear spectroscopy and imaging, opening a pathway to gain direct insight into the coupled motions of electrons and nuclei with resolution on the electronic length scale and timescale. Zhang et al. have proposed and successfully generated such innovative X-ray pulse pairs using a nonlinear optical process known as superfluorescence [7]. Their concept involves utilizing a tightly focused XFEL pulse with pronounced double spikes in the temporal profile, which is directed at a thin metal foil with an absorption edge situated below the X-ray photon energy. As coherent fluorescence emission selectively arises from these two strong spikes, it becomes feasible to produce phase-stable X-ray pulse pairs. This novel X-ray source has the potential to serve as a foundation for frequency combs in the hard X-ray region.

4-3. Single shot observation of ultrafast surface nanodynamics

Detailed understanding and control of the first interactions of an intense laser and matter are expected to enable precise material processing with lasers and high-energy-density science. The research group led by Randolph et al. conducted an experiment to observe the ablation and the density dynamics of the surface of laser-irradiated matter for the first time with picosecond temporal and nanometer depth resolution [8]. They irradiated a high-power femtosecond laser onto metallic multilayer targets to cause surface ablation and density perturbation. Then, the surface was irradiated by an XFEL pulse at a grazing-incidence angle with a variable delay. The single-shot GIXAXS (grazing-incidence small-angle X-ray scattering) pattern is used to reconstruct the density profiles of the multilayer targets at the probed timing. The results will help further improve the simulation capabilities in laser–matter interactions.

4-4. XFEL imaging for intact sea-island nanostructures of solid-state electrolytes

Suzuki et al. observed sea-island nanostructures of solid-state electrolytes without irradiation damage [9]. They used a novel high-contrast coherent diffractive X-ray imaging technique that quantitatively analyzes the intact internal structure of metastable glass−ceramic nanoparticles using femtosecond XFEL pulses. The volume ratio of the amorphous to crystalline phases for the measured metastable sulfide glass−ceramic particle $[(Li_2S)_{70}-(P_2S_5)_{30}]$, which is utilized as electrolytes for all-solid-state batteries, was evaluated to be ~2.5:1. Their results are expected to facilitate structural studies on fragile metastable materials of scientific and industrial importance.

Inoue Ichiro[*1], Inubushi Yuichi[*1,2], Kang Jungmin[*1], Miyanishi Kohei[*1], Osaka Taito[*1], Owada Shigeki[*1,2], Yamaguchi Gota[*1], Yabuuchi Toshinori[*1,2], Tono Kensuke[*1,2], and Yabashi Makina[*1,2]

[*1] Advanced Photon Technology Division, RIKEN SPring-8 Center

[*2] XFEL Utilization Division, Japan Synchrotron Radiation Research Institute

References:

[1] Iwayama, H. et al. (2020). *Appl. Sci.* **10**, 7852.

[2] Inoue, I. et al. (2019). *Nat. Photonics* **13**, 319.

[3] Yumoto, H. et al. (2012). *Nat. Photonics* **7**, 43.

[4] Osaka, T. et al. (2022). *Phys. Rev. Res.* **4**, L012035.

[5] Yumoto, H. et al. (2022). *Nat. Commun.* **13**, 5300.

[6] Maestre-Reyna, M. et al. (2022). *Nat. Chem.* **14**, 677.

[7] Zhang, Y. et al. (2022). *Proc. Natl. Acad. Sci. USA* **119**, e2119616119.

[8] Randolph, L. et al. (2022). *Phys. Rev. Res.* **4**, 033038.

[9] Suzuki, A. et al. (2022). *Nano Lett.* **22**, 4603.