4-4. SACLA Beamlines

1. Operation status

In FY2023, SACLA was operated stably for over 6,000 hours, which was consistent with the past few years after the COVID-19 pandemic, for user experiments at the three beamlines in total. Note that one of the three beamlines, BL1, is a soft X-ray (SX) free-electron laser (FEL) beamline and the other two beamlines, BL2 and BL3, are hard X-ray FEL (XFEL) beamlines.

The major topics on SACLA's beamlines and experimental capabilities in this fiscal year are summarized in the following subsections. Some of them were the achievements attained through the SACLA/SPring-8 Basic Development Program. Close collaborative activities with external experts under this program enable the prompt realization of efficient and effective developments.

2. SX-FEL beamline (BL1)

Two nanofocusing systems are currently under development with the support of the Basic Development Program. The first is a general-purpose system designed to stably produce a small SX-FEL beam approximately 500 nm in diameter. The second, called the wavelength-scale focusing system, aims to achieve beam diameters as small as 50 nm using the focusing optics with a high numerical aperture (NA).

The general-purpose system utilizes a Wolter type-I mirror with a full-circumference shape. The mirror fabricated through nickel electroforming has a reflectivity of about 50% at both the fundamental and third harmonics of BL1. It also has a high reflectivity in the infrared region. This system accepts a relatively large angular alignment tolerance, thus ensuring stable submicron focusing. The commissioning of focusing using the two-color beams, i.e., the SX-FEL and the synchronized optical laser, was started in FY2023 for prospective applications in time-resolved pump-probe experiments.

The wavelength-scale focusing system consists of a ring-focusing mirror and a quasi-ellipsoidal mirror. Although the development of this system is progressing, achieving diffraction-limited focus remains a challenge because of the required extreme preciseness of mirrors. In FY2024, the refinements of the mirror fabrication techniques are planned to improve the mirror quality.

3. XFEL beamlines (BL2 and BL3)

3-1. Pulsed magnetic field generation system over 100 Tesla

Magnetic fields are one of the fundamental external fields in condensed matter physics. Recently, various novel and exotic states of matter have been reported under magnetic field strengths in the vicinity of 100 T. Electromagnets can produce such high magnetic fields but the fields are produced in short lifetimes (μs to ms) and at low repetition rates (once every tens of minutes). XFELs are powerful for obtaining microscopic information on materials under such magnetic fields in a single-shot experiment.

At SACLA, a portable pulsed power system was developed under the Basic Development Program and generated a 120 T magnetic field in FY2023. A demonstration experiment of X-ray diffraction was conducted with this high magnetic field, resulting in a successful observation of a structural phase transition in a strongly correlated material. The system still has challenges regarding the stability of magnetic field generation, which is expected to be addressed soon.

3-2. Drop-on-tape sample delivery system for serial femtosecond crystallography at SACLA

Time-resolved serial femtosecond crystallography (SFX) is one of the most reliable methods for observing the structural dynamics of proteins under physiological conditions. However, this method has some challenges regarding the large amount of sample consumption and the excitation reaction of non-photosensitive proteins.

A drop-on-tape sample delivery system has the potential to overcome the above challenges. A prototype of the system has been developed at SACLA. As Fig. 1 shows, the system employs two piezo-type nanoliter droplet dispensers and a reel-to-reel tape transfer. The sample droplets attached to a thin polyimide film tape are vertically transported and delivered to the XFEL pulses with high reliability. The mix-and-inject SFX experiment for the enzymatic binding reaction has been successfully demonstrated by directly mixing two droplets on the tape maintaining a sample consumption close to that for the high-viscosity injector. Further developments to reduce the droplet volume and fulfill the observation of the sub-second reaction remain.

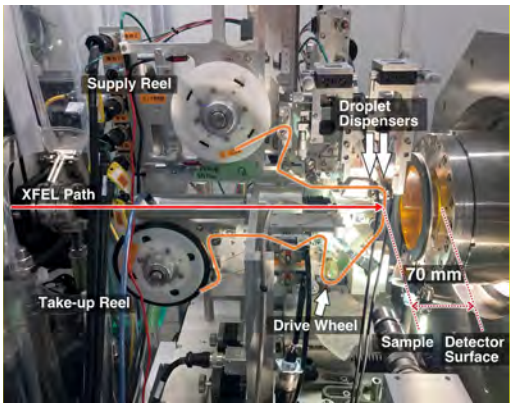

Fig 1. Drop-on-demand sample delivery system integrated into the SFX platform.

3-3. Sample exchange system for high-intensity laser experimental platform

Experiments using a high-intensity optical laser in EH6 are conducted in a large vacuum chamber. The number of samples contained in the chamber, currently around 100–150, limits the shot numbers because the sample is destroyed by laser irradiation. Once all samples are shot, the chamber is opened, new samples are installed, and the chamber is pumped down. The process takes more than a few hours and is followed by a realignment of the laser optics before data acquisition resumes. Sample replacement typically occurs every night during beamtime.

An in-vacuum sample exchange system has been developed recently. A more than three times larger number of samples can be contained than the current one. The new system improved experiment efficiency and data reproducibility significantly. The sample exchange system is under commissioning for the first use in FY2024.

4. Synchronized Optical Lasers

4-1. Accessibility improvements of synchronized femtosecond laser systems

Two sets of millijoule-class Ti:sapphire laser systems have been utilized in ultrafast pump and probe experiments at SACLA. The two laser systems had been located in LH1 and operated for independent experiments in parallel, one at BL1 and the other at BL3, until FY2022. Because of the large demand for the pump-probe experiments, it has been decided to build a new laser hutch, LH2, and to isolate the laser systems to reserve sufficient maintenance and optimization time.

LH2 was constructed in FY2022 and one laser system was rehoused in LH2 during the summer shutdown in 2023. The laser system immediately restarted its user operation for experiments at BL1 on schedule. From FY2024, this laser system also delivers the beam to EH3 at BL2, which expands the opportunities for ultrafast pump and probe experiments.

4-2. Few-cycle pulse capabilities in synchronized optical laser

The typical pulse duration of the synchronized femtosecond laser pulses is 40 fs, which is one of the factors limiting the temporal resolution to observe ultrafast phenomena. A shorter pulse laser is desired to achieve higher temporal resolution; therefore, a system to produce a few-cycle pulse has been developed. The system is based on bandwidth broadening through a self-phase modulation in a gas-filled hollow-core fiber followed by pulse compression using chirped mirrors. The output pulse from the system has been characterized to have a duration of 10 fs and energy of 200 μJ. The few-cycle pulses have been in trial use at BL1 in FY2023 but will be fully utilized in user experiments after FY2024 at both BL1 and BL3.

4-3. Development of terahertz-pump capabilities for X-ray-probe experiments

A nearly single-cycle terahertz pulse with a large electric-field amplitude has been applied to control the physical properties of various solids. The low photon energy of the terahertz pulse is suitable for exciting quasiparticles and studying ultrafast quantum phase transitions. A sub-cycle pump-and-probe spectroscopy system using intense terahertz pulses was designed and implemented in the X-ray diffractometer in FY2023 to extend the optical pump capabilities mainly for materials science. By using parabolic mirror scaling optics, a focal diameter of about 1 mm of the terahertz pulse has been achieved. After estimating the electric-field amplitude of the terahertz pulse, a demonstration of pump-and-probe spectroscopy will be carried out. This measurement system is anticipated to be ready for use in FY2024.

5. Research highlights

5-1. Fast structural dynamics of Photosystem II during the water oxidation process

Photosystem II (PSII) is the protein complex that catalyzes the oxidation of water to produce hydrogen ions and molecular oxygen in photosynthesis. Li et al. have investigated the light-induced dynamics of PSII using time-resolved serial femtosecond crystallography on nanosecond and microsecond timescales [1]. They observed concerted movements of water molecules and surrounding amino acid residues, which would promote the sequence of electron transfer, proton release, and delivery of a substrate water molecule into the catalytic center for molecular oxygen formation. These results will provide useful information for the development of artificial photosynthesis systems.

5-2. 7 nm focusing of XFEL pulses achieving 10^{22} $\mathrm{Wcm^{-2}}$ intensity

High-intensity X-ray fields achieved through the extreme focusing of XFEL pulses have ushered in a new era in X-ray nonlinear optics and the study of intense X-ray and matter interactions. Yamada et al. have surpassed previous X-ray intensity records by two orders of magnitude, achieving an intensity beyond 10^{22} Wcm^{-2} [2]. They employed a new focusing system based on the Wolter-III geometry, realizing a spot size of 7 × 7 nm^2 for 9.1 keV XFEL pulses, with an operational lifetime of over 10 h. This unprecedented intensity should give rise to novel opportunities for exploring atomic, molecular, and optical physics, plasma physics, and X-ray nonlinear optics. Moreover, it could advance single-molecule imaging for biomolecules.

5-3. Capturing transonic dislocation propagation in diamond

The motion of dislocations has been extensively studied for decades because it plays a key role in plastic deformation. However, how fast the dislocations propagate has been poorly understood experimentally. Katagiri et al. have tracked ultrafast dislocation motion in single-crystal diamonds by femtosecond X-ray radiography in laser-shocked single-crystal diamonds [3]. The captured images of the deformation time evolution reveal, for the first time, that the dislocation propagation speed in shocked diamond exceeds its transverse sound speed. This observation is crucial for refining models that provide insights into ultrafast deformation behaviors, including ultrafast fracture in structural materials and earthquake ruptures.

5-4. Tracking nuclear motion in single-molecule magnets

Single-molecule magnets (SMMs) are at the forefront of research in fields such as quantum computing, spintronics, and high-density data storage, owing to their ability to retain magnetic information at the molecular level. However, understanding their behavior on ultrafast timescales, where nuclear and electronic dynamics interplay, remains a significant challenge. Barlow et al conducted femtosecond time-resolved X-ray absorption spectroscopy (TR-XAS) and revealed that this molecule is rigid, as expected from the molecular design, and changes in its nuclear structure alone will be unlikely to achieve the control of magnetization using photons [4]. Femtosecond TR-XAS can shed light on these small geometry changes and provides pivotal guidelines to control the magnetization in SMMs using light.

YABUUCHI Toshinori[*1,2], KATAYAMA Tetsuo[*1,2], KANG Jungmin[*1], KIDA Noriaki[*1,2], KUBOTA Yuya[*1], MIYANISHI Kohei[*1], OSAKA Taito[*1], OWADA Shigeki[*1,2], TOGASHI Tadashi[*1,2], TONO Kensuke[*1,2], YAMAGUCHI Gota[*1], 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] Li, H. et al. (2024). *Nature* **626**, 670–677.
[2] Yamada, J. et al. (2024). *Nat. Photonics* **18**, 685.
[3] Katagiri, K. et al. (2023). *Science* **382**, 69.
[4] Barlow, K. et al. (2024). *Nat. Commun.* **15**, 4043.