

## 4-4. SACLA Beamlines

### 1. Operation status

All three beamlines at SACLA were stably operated through FY2024. The total operation time for users was of the same order as the past few years, which was over 6,000 h. Approximately 100 user experiments were conducted this fiscal year at the soft X-ray free-electron laser (FEL) beamline (BL1) and the two hard X-ray FEL beamlines (BL2 and BL3). In addition to user operations, SACLA has initiated R&D efforts to generate attosecond hard X-ray pulses at BL2, using the self-chirping scheme. In FY2024, pulse shortening was observed down to  $\sim 2$  fs [full width at half maximum (FWHM)], indicating the effectiveness of the self-chirping approach at SACLA as a preliminary trial. Additional studies are scheduled for the next fiscal year to establish schemes for producing and characterizing attosecond X-ray pulses.

The other specific achievements and ongoing projects on beamlines and experimental capabilities are outlined below.

### 2. Updates on Experimental Capabilities

#### 2-1. Two-dimensional large-area detector

Serial femtosecond crystallography (SFX) has become one of the most popular techniques for determining macromolecular structures using XFEL. Recently, the objects of SFX have been expanded to small molecules, which demand subangstrom resolution and thus a detector with both a large imaging area and a short working distance (WD). We have implemented a large-area detector, comprising 72 tiled sensor modules of CITIUS. The detector, CITIUS-20.2M, has an imaging area of  $\sim 32$  cm in the horizontal direction

and  $\sim 39$  cm in the vertical direction. The detector can be placed at a minimum WD of 70 mm on a diffraction platform at SACLA for enabling reflection angles up to  $2\theta \sim 70^\circ$ , corresponding to a resolution of  $0.7 \text{ \AA}$  or higher at 15 keV. A diffraction image-sorting pipeline for the SFX experiments has been rebuilt for the new data format output from CITIUS-20.2M. A demonstration SFX experiment on lysozyme microcrystals was performed at EH3 on BL2. Pilot experiments for small-molecule SFX at photon energies above 15 keV were also conducted in this fiscal year.

#### 2-2. Portable mirror system for nano-focusing

A portable nano-focusing system utilizing Kirkpatrick–Baez (KB) optics was developed<sup>[1]</sup> to enable the integrated use of nano-focused XFELs with experimental instruments, such as a large-area detector and an optical laser system for pump–probe experiments. The newly developed system has a total length of only 340 mm from the first elliptical mirror to the focal point, as shown in Fig. 1. The large glancing angle of 5 mrad enables a spatial acceptance of  $650 \mu\text{m}$ , even for the small effective mirror length of 130 mm. During the demonstration

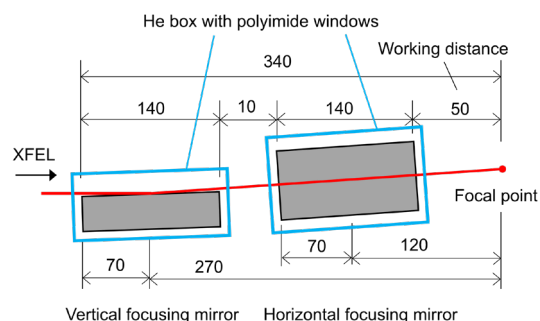


Fig. 1. Schematic image of the side view of the nano-focusing optics. The unit of length is millimeters.

of this system, a focal spot as small as  $150 \text{ nm} \times 220 \text{ nm}$  (FWHM) was achieved, resulting in a focused intensity of  $2.5 \times 10^{19} \text{ W/cm}^2$  for 10 keV XFEL pulses with a pulse energy of 110  $\mu\text{J}$ . The system has been in use at BL2 EH3 and BL3 EH2/4c since the 2024A term.

### **2-3. Stabilization of pulsed magnetic field generation system over 100 Tesla**

In condensed matter physics, numerous novel and exotic states of matter have been reported under magnetic fields of around 100 T. To advance such studies, a portable pulsed-power system was developed under the Basic Development Program, and a magnetic field strength of 120 T was achieved in FY2023. Microscopic lattice structures of exotic states in high magnetic fields can be directly probed with this system at SACLA. The system initially exhibited instability in magnetic field generation, primarily owing to fluctuations in the airgap switches that are responsible for delivering high currents to the electromagnet. In FY2024, this issue was resolved by optimizing air control in the switches, thereby stabilizing the field generation. This advancement enabled the acquisition of high-quality X-ray diffraction data in high magnetic fields for user experiments.

### **2-4. Integration of pulsed magnetic field generation system into high-power nanosecond laser platform**

Applying a pulsed magnetic field to a sample in high-power laser experiments advances the exploration of various physical phenomena in astrophysics, planetary science, and inertial confinement fusion. Within the Basic Development Program, a pulsed magnetic field generation system

exceeding 10 T has been developed for a high-power nanosecond laser platform at SACLA. XFELs can precisely investigate the dense plasmas and condensed matter in such a system. The system has been integrated and tested, demonstrating stable magnetic field generation under simultaneous nanosecond-laser irradiation. A demonstration experiment of XFEL imaging under high magnetic fields is planned, along with further development to increase the maximum achievable field strength.

### **2-5. Automatic sample exchange system in vacuum for high-power laser experiments**

At the high-power femtosecond laser platform, a new sample must be repositioned at the interaction point in vacuum after each laser shot, as the shot destroys the sample. As a result, the number of shots is limited by the number of samples loaded in the chamber, typically 100–150 per cycle. To avoid several hours of evacuation and optical realignment, it has been desired to increase the number of stored samples and enable sample exchange without breaking the vacuum. An in-vacuum sample exchange system was developed and first applied in user experiments in 2024B. This system can accommodate more than three times the number of samples compared with the previous setup in a single pumping cycle. It has improved operational efficiency and data reproducibility by reducing condition changes associated with chamber purges.

## **3. Updates on Synchronized Optical Lasers**

### **3-1. Synchronized optical laser systems**

Femtosecond pulse lasers with temporal durations of several tens of femtoseconds are widely employed in ultrafast pump–probe experiments. At SACLA, two millijoule-class Ti:sapphire laser

systems are available for experiments. The two laser systems have been operated in separate laser hutches (LH1 and LH2) to allow sufficient time for maintenance and optimization since FY2023. The long-term timing stability has been improved for the LH1 system dedicated to femtosecond pump–probe experiments at BL3 [2]. In addition, the LH2 system has started supplying the beam to EH3 at BL2 in addition to EH4a at BL1 from the term 2024A. To date, the lasers at EH3 are applied to pump–probe experiments that require picosecond temporal resolution. Femtosecond pump–probe capabilities will also be available at EH3 after the commissioning of a timing monitor system scheduled in FY2026.

### 3-2. New capabilities in pulse duration and in wavelength of synchronized lasers

We have been working to expand pump capabilities with the synchronized laser systems. One direction is to shorten the pulse. Few-cycle pulses in the near-infrared region were fully utilized starting from FY2024. The system is based on bandwidth broadening through a self-phase modulation in an argon gas-filled hollow-core fiber, followed by pulse compression using chirped mirrors. The typical pulse duration is 10 fs (FWHM), with a pulse energy of 200  $\mu\text{J}/\text{pulse}$  and a focal spot size of 100  $\mu\text{m}$  (FWHM), using a silver-coated concave mirror with a focal length of 1000 mm. The pulses are available at BL1 (EH4a) and BL3 (EH2).

Another direction is to expand the range of available wavelengths. A subcycle pump-and-probe spectroscopy system utilizing intense terahertz pulses was designed and implemented in the X-ray diffractometer, primarily for materials science applications. The low photon energy of the terahertz

pulse is suitable for exciting quasiparticles and studying ultrafast quantum phase transitions. By using nonlinear organic crystals, we successfully detected the emitted electric field waveforms of terahertz pulses and achieved a strong electric field of about 0.4 MV/cm and a center frequency of about 2 THz. This terahertz pump and X-ray diffraction probe measurement system has been in operation for user experiments since the 2024B term.

## 4. Research Highlights

### 4-1. Ultrafast dual-pulse control of phase transition in $\text{VO}_2$

Vanadium dioxide ( $\text{VO}_2$ ) is one of the model materials for studying photoinduced insulator-to-metal transitions, where both structural and electronic changes are closely linked. Ultrafast studies reveal that its structural and electronic dynamics are closely intertwined, yet the transition mechanisms remain unclear. Johnson et al. reported a dual-pulse excitation that utilizes a weak prepulse to induce structural fluctuations, thereby lowering the energy barrier for the transition [3]. This challenges the traditional emphasis on coherent phonons by showing that disorder can play a central role. Their findings provide new insights into ultrafast scattering techniques and the energy-efficient control of phase transformations.

### 4-2. Ultrafast photoinduced insulator–metal transition in a Mott insulator

Photoinduced insulator–metal transitions in Mott insulators are attracting attention for their potential in ultrafast switching. However, the electronic transition is accompanied by the nucleation and growth of percolating domains of the changed lattice structure, leading to empirical timescales

dominated by slowly coarsening dynamics. Verma et al. performed time-resolved X-ray diffraction and reflectivity measurements [4]. They revealed that the transition in an epitaxially strained Mott insulating thin film occurs without domain formation or coarsening effects, thereby exposing the intrinsic dynamics. Their findings underscore the need to investigate both electronic and structural aspects in photoinduced phenomena.

#### 4-3. Heating dynamics of matter irradiated by intense laser pulses

Interactions of high-intensity, short-pulse lasers with matter generate energetic electrons and create a dense plasma. However, diagnosing the dynamics of laser-matter interactions has been challenging because of their highly transient nature. Sawada et al. achieved submicron- and femtosecond-resolution measurements of a solid-density copper foil heated by laser-driven fast electrons [5]. Observed X-ray transmission images revealed a formation of hot plasma in the laser spot region, surrounded by warm dense matter. They also captured images showing energy relaxation processes occurring over tens of picoseconds. These findings validate advanced simulations and provide new insights into energy transport, advancing research in high-energy-density science.

#### 4-4. Observation of living mammalian cells by femtosecond soft X-ray microscopy

Soft X-ray microscopy provides powerful contrast for cellular imaging; however, radiation damage has limited its application in living cells. Egawa et al. developed a soft X-ray microscope using femtosecond pulses from a free-electron laser and Wolter mirrors, enabling the single-shot imaging of

living cells with negligible damage [6]. They successfully visualized mammalian living cells in culture medium, revealing carbon distributions such as nucleoli and nuclear membranes. The broad wavelength tunability of soft X-ray free-electron lasers, combined with the achromaticity of Wolter mirrors, enabled wavelength-resolved cellular imaging. This work demonstrates that femtosecond soft X-ray microscopy can capture structural features of living mammalian cells previously inaccessible by conventional methods.

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