How Scientists at SACLA Enhanced Their Pump-Probe Experiments with Precise Synchronization

In 2020, a group of scientists around Dr. Makina Yabashi and Dr. Tadashi Togashi (SPring-8 Center, RIKEN) introduced a new femtosecond optical laser system for pump-probe experiments at SPring-8 Angstrom Compact free-electron Laser (SACLA) in Harima, Japan. On its first light in 2012, SACLA was the world's second hard x-ray free-electron laser (XFEL), marking a new era in X-ray photonics. Since then, it has been a place where scientists push the borders of our knowledge every day and try to gain fundamental insights on how our world works.

Figure 1 SACLA at Spring-8, RIKEN (Photo copyright RIKEN, http://xfel.riken.jp/)

Dr. Tadashi Togashi is an expert on time-resolved x-ray measurements using femtosecond-lasers at the SACLA facility of SPring-8. Besides his own research, he is responsible for the optimization of the laser systems for user experiments. Over the past 20 years, he has contributed to over 350 peer-reviewed publications and with his latest project, he can again improve the results of many other scientists working at SACLA.

In this project, Dr. Togashi optimized SACLA's laser setup for pump-probe experiments. The pump-probe setup allows time-resolved x-ray experiments and provides insights into chemical, electronic and structural transitions in molecules or other samples. At SACLA, a femtosecond optical laser excites these transitions – it "pumps" the sample into an excited state. After a given time, the sample relaxes again. To monitor this relaxation process, the sample is "probed" with x-ray pulses in a series of measurements while the time interval between the pump and the probe pulse changes for each measurement. This leads to a series of diffraction patterns or spectra which creates a movie on the molecular level.

In principle, the short pulses of the free-electron laser allow for femtosecond time resolution. However, the resolution is in practice often limited to >100 femtoseconds because of synchronization issues or jitter between the femtosecond laser's pump pulses and the x-ray probe pulses. This was also the case at SACLA.

Since 2013 SACLA was operating their pump-probe setup using the XFEL's x-ray pulses [1]. The pump laser was a Ti:sapphire (TiS) chirped pulse amplification system (CPA, Legend Elite, Coherent Inc.) consisting of a mode-locked TiS oscillator (Micra, Coherent Inc.) followed by an amplifier. The amplified pulses were used to drive an optical parametric amplifier (OPA, OPerA Solo, Coherent Inc.) or BBO crystals for wavelength conversion (see Figure 2).

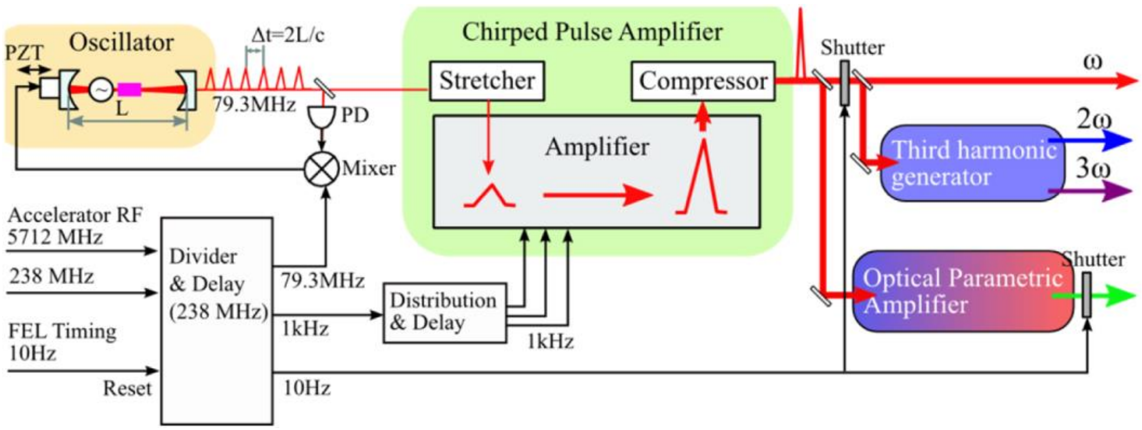

Figure 2 Former laser system at SACLA using a timing approach based on direct photodetection. (from [2])

As mentioned before, pump-probe experiments rely on an accurate synchronization between the femtosecond-pump-laser and the x-ray probe-pulses of the FEL. Therefore, all systems have to work on a shared time base – in this case, the master clock of the SACLA accelerator. For this reason, it is necessary to adjust the TiS laser oscillator's internal frequency to match the external master, which initially was done with a Coherent Synchrolock AP.

The Synchrolock monitors a fraction of the TiS oscillator's light and detects the laser's repetition rate using direct photodetection. The repetition rate is then electronically compared with the reference signal from the SACLA master clock. Based on this signal, the TiS oscillator's cavity length is changed. This results in an adjusted oscillator frequency and improves the timing of subsequent pulses.

However, this electronic synchronization system left a residual jitter of approx. 300 femtoseconds which limited the overall temporal resolution of the experiments at SACLA. Since many time-resolved X-ray measurements need a very fine time resolution and time delay step-size between pump and probe pulses, the electronic approach with direct photodetection was reaching its performance limits here.

Faced with this challenge, Dr. Togashi and his colleagues focused on several aspects that influenced the stability and accuracy of the setup. They optimized subsystems like the OPA (changed to a TOPAS HE-PRIME, Light Conversion, Vilnius, Lithuania) or an "acousto-optic programable dispersive filter (AOPDF)" (Dazzler, FASTLITE, Antibes, France). This helped to improve the dispersion in the optical path. Also, the team put strong emphasis on minimizing external interferences like temperature variations, vibrations and airflow. However, the synchronization of the laser systems with the SACLA master clock remained challenging. To improve the timing jitter of their new pump-probe setup, the team around Dr. Togashi had to optimize the synchronization of the TiS oscillator to the master clock.

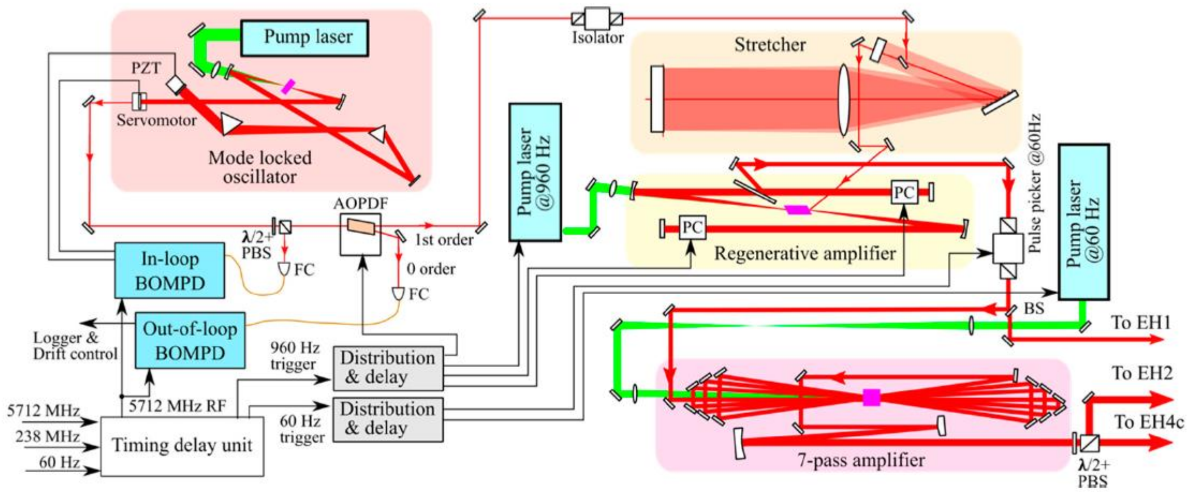

Figure 3 New laser design at SACLA with a synchronization based on a Balanced Optical Microwave Phase Detector. (from [3])

So, when Dr. Togashi heard about the Cycle Balanced Optical Microwave Phase Detector (BOMPD) from colleague at SACLA's accelerator and timing team, he instantly knew that this device could help him with his synchronization issues. The fully-automated BOMPD precisely detects the time delay between an optical pulse train and the zero-crossings of a microwave signal. It generates a signal that is proportional to the timing error between the two inputs and is used in a phase locked loop configuration to tightly synchronize a laser to a microwave source. Due to its balanced detection scheme, the BOMPD is immune to amplitude fluctuations of both optical and microwave sources and greatly suppresses AM-PM conversion noise in the photodetection process.

Figure 4 Cycle BOMPD, a fully-automated device to precisely lock lasers and RF sources

"The existing Synchrolock System is mainly based on electronic devices to synchronize the system, which limits the timing accuracy of the complete system" explains Dr. Togashi. "The BOMPD in contrast offers detection of the phase error in the optical regime. This significantly enhances the synchronization accuracy. And it is possible to lock the TiS oscillator directly to the 5.7 GHz RF signal of the accelerator's master clock without changing the signal."

After receiving the BOMPD (Cycle GmbH, Germany), the scientists installed the new synchronization (see Figure 3) and had their test setup running within three weeks – and with exciting results: Cycle's approach allowed to significantly improve the synchronization accuracy. Now, after transferring the configuration to the XFEL beamline, the complete pump-probe system shows a timing jitter of 47 femtoseconds compared to > 300 femtoseconds before optimization – a reduction by almost an order of magnitude. This result was confirmed using a relative arrival time monitor between the XFEL pulses and the optical laser pulses (see Figure 5a and b).

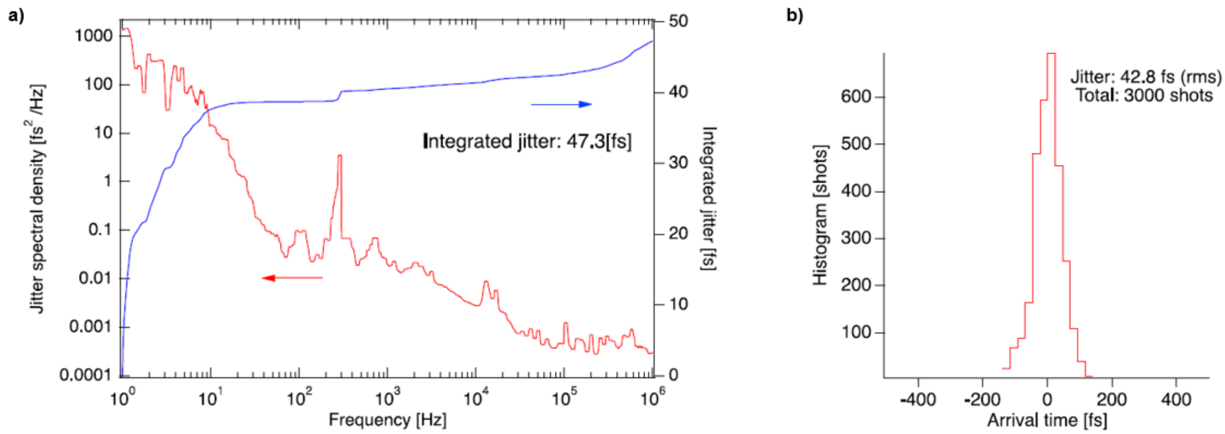

Figure 5 Timing jitter of the new SACLA synchronization system using a BOMPD. a) Measurement of the Pump-probe setups integrated timing jitter b) Measurement of the timing jitter using a beam arrival time monitor. A histogram of the difference of pulse arrival over 3000 shots is shown. (adapted from [3])

In a publication from November 2020, Dr. Togashi presented the new setup and showed first experimental results. As a reference experiment, the SACLA team measured time-resolved X-ray diffraction (tr-XRD) of photo-excited bismuth (Bi). Its coherent phonon oscillation has been a standard evaluation experiment for pump-probe measurements using XFEL and femtosecond optical laser.

In the experiments, the femtosecond laser excites the bismuth crystal, and the intensity of the diffracted X-rays is measured with a 33-femtosecond step size. When using the old photodetection-based synchronization system, its 300-femtoseconds jitter blurred the phonon signal, and the scientists needed an additional timing monitor for jitter error correction to reveal the signal from the noise (see Figure 6a). When the same experiment is measured using the BOMPD for synchronization, even in the unprocessed raw data the coherent phonons were easily observed (see Figure 6b). The raw data is already so good that further post-processing with the timing monitor does not reveal additional information. Dr. Togashi summarizes: "The BOMPD synchronization offers high timing performance without an additional timing monitor and without the big effort of post-processing, which is not always possible. And it also offers a much better control over the timing step-size of our time resolved measurement when compared to the former synchronization system."

With these improvements, the new pump-probe setup at SACLA now offers a highly stable laser system with an overall jitter below 50 femtoseconds. The setup is ready for the upcoming challenges of high precision time-resolved X-ray experiments. And thanks to the work of Dr. Togashi and his colleagues, scientists from around the world can come to SACLA and continue to achieve unique insights into the ultrafast dynamics of their samples.

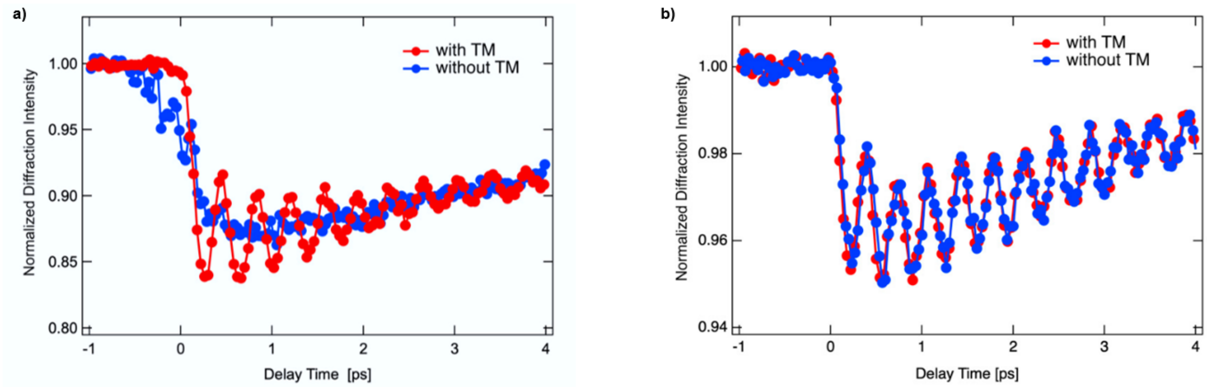

Figure 6 Diffraction intensity of Bi (111) as a function of delay time. The optical laser was synchronized using a fast photodetector (a) and using a BOMPD (b). The blue and red traces show the raw data obtained by scanning the delay and the data post-processed using the timing monitor (TM). The measurements with the BOMPD directly detect the coherent phonon oscillation of the photo-excited Bi without the need for laborious post-processing with the additional timing monitor (adapted from [3])

Cycle contact

For more information on Cycle's BOMPD as well as other timing and synchronization products or ultrafast fiber lasers, visit cyclelasers.com or email sales@cyclelasers.com.

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