（別紙様式５）

変革を駆動する先端物理・数学プログラム（FoPM）
国外連携機関長期研修　報告書

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Under the supervision of Professor Craig P. Schwartz in University of Nevada, Las Vegas, I made my research trips to participate in experiments of nonlinear X-ray spectroscopy at the two facilities of X-ray Free Electron Laser (XFEL), the EIS-TIMER beamline at Free Electron laser Radiation for Multidisciplinary Investigations (FERMI) at Trieste city in Italy and at SXFEL beamline at SPring-8 Angstrom Compact free-electron LAser (SACLA) at Hyogo prefecture in Japan.

Nonlinear optical responses in matters have been one of the frontier science topics in various research fields due to observations of the optical phenomena that cannot be described by conventional theories of the linear response. The second-order non-linear optical responses, such as second harmonic generation (SHG), are unique phenomena that double the frequency of light (from hν to 2hν) and occur at a system with the broken inversion symmetry, such as at surfaces/interfaces. While the up-conversion mechanism itself has captured interests among researchers, the SHG experiment has been also used as a measurement method to selectivity probe surfaces/interfaces. The construction of XFEL has extended wavelengths (λ's) of the optical SHG experiments to the soft X-ray region (λ~10 nm) that can satisfy the inner-shell excitation. Such a core-level resonance or element-selectivity of SHG was observed under reflection geometry at SACLA [Sh. Yamamoto *et al.*, Phys. Rev. Lett. **120**, 223902 (2018).] and under transmission geometry at FERMI [R. K. Lam *et al.*, Phys. Rev. Lett. **120**, 023901 (2018).]. Up to now, the soft X-ray SHG measurements have transferred from a proof-of-principles instrument to a spectroscopy for materials science, and the effect has been reported from materials, such as magnetic multilayer-structured samples [T. Sumi *et al.*, Appl. Phys. Lett. **122**, 171601 (2023).], semiconductors [T. Sumi *et al.*, e-J. Surf. Sci. Nanotech. **20**, 31 (2022).], solar cells [M. Horio *et al.*, Appl. Phys. Lett. **123**, 031602 (2023).]. Under the current program of my research trip, I participated in beamtime of such soft-Xray SHG experiments: (i) Observation of newly non-linear optical effect in soft X-rays and (ii) Investigation of soft X-ray SHG originating from buried interfaces in solar cells.

(i) Observation of newly non-linear optical effects in soft X-rays

In this experiment, we challenged to observe sum frequency generation (SFG) in soft X-rays for the first time in the history of science at at EIS-TIMER beamline at FERMI. The sample was a MoS_2 wafer, which is one of the transition metal dichalcogenides. Using the high-harmonics in the incident XFEL beam, two types of lights were used : $h\nu_1 = 99.4$ eV and $h\nu_2 = 132.5$ eV. The beams were separated by a transmission-typed grating and then, irradiated onto the sample spatiotemporally to induce the SFG light at $h\nu_1 + h\nu_2 = 231.9$ eV satisfied the inner shell resonance with the Mo *M*-shell absoprion edges. The SFG signals were observed by photodiodes which were placed at transmission and reflection positions. Although much data are needed to justify the result, we accumulated the skill of the detection for the future experiment.

During the beamtime, I walked from my room to FERMI beamline every day. The air was very clear because the synchrotron radiation facility was surrounded by grassland. One of the days was the Italian National Day and I enjoyed the atmosphere of the country. Before going back to Japan, I visited Miramare Castle (Castello di Miramare in Italian), which is a castle built by Maximilian I near Trieste. I watched the beautiful sight of the castle with Gulf of Trieste. Besides the research, the experiences were also valuable.

Figure. Photos of the optical setup for soft X-ray SFG we constructed at EIS-TIMER beamline at FERMI, taken at (left) the upstream and (right) the downstream of the XFEL beam.

(ii) Investigation of soft X-ray SHG originating from buried interfaces in solar cells

In this SHG experiment, we aimed to investigate electronic states at buried interfaces in solar cells that is responsible for the functionality. This experiment was performed at SXFEL beamline at SACLA. The incident light was irradiated onto a sample with an angle of 45°. The reflected light was guided to a reflection-typed grating to separate the fundamental and SHG lights, followed by the observation by a two-dimensional detector [Sh. Yamamoto *et al.*, Phys. Rev. Lett. **120**, 223902 (2018).]. The energy of the incident light was set to hν ~ 100 eV to satisfy the resonance effect with the Si *N*-shell absorption edges in SiO_2 [M. Horio *et al.*, Appl. Phys. Lett. **123**, 031602 (2023).]. Through the measurement of the various solar cell samples, we detected the SHG spectra of the buried SiO_2 interface. The theoretical calculation has now been conducted to understand difference in the appearance of SHG from the samples.

In these trips, I experienced a lot in the beamtimes. These splendid experiences were invaluable in my life as a researcher. Furthermore, I felt my English skills, especially speaking and listening abilities, were improved through discussions with other researchers on these trips. Finally, I would thank Professor Craig Schwartz, Dr. Cristina Bömer, and other researchers for allowing me to participate in these beamtimes and Professor Iwao Matsuda for introducing them.