

Development of a diamond X-ray phase retarder to generate a circularly polarized XFEL beam at SACLA

An X-ray free electron laser (XFEL) is a light source that combines the unique features of laser and X-rays. XFEL radiation is characterized by extremely high brilliant, transversely coherent, and ultrashort X-ray pulses. Using this probe with unprecedented capabilities, atomic-scale observation of ultrafast dynamics in materials and biological systems would be feasible. XFEL could offer potential applications that have yet to be explored using storage-ringbased synchrotron radiation. One unexploited feature of XFEL is X-ray polarization. This is in contrast to variable X-ray polarization, which has been a standard technique used in various applications at storage ring sources.

Generally, XFEL sources produce a linearly polarized electromagnetic wave, in which the electric field vector oscillates in the horizontal plane. However, researchers have desired XFEL beams in other polarization states where the electric field vector is rotating spirally (i.e., circular polarization). Circularly polarized XFEL can extend the range of SACLA's applications, making it possible to observe electron spin motions in magnetic materials and structural dynamics of chiral materials in biological systems in a time scale shorter than 1 picosecond. Full utilization of the XFEL polarization property should expand the capability of this powerful light source.

In this study, we developed an instrument to control the polarization states of an XFEL beam [1]. Figure 1 shows a schematic of our X-ray polarizer optics, which is similar to the system established at the beamlines of storage ring sources [2]. A synthetic diamond crystal was used as a transmission-type phase retarder. The diffraction plane is tilted 45° from the direction of incident linear polarization, and the crystal angle is set at near the Bragg condition. As X-rays pass through the crystal, a phase shift between the σ - and π -polarization components is generated, which is controllable with the crystal angle. At an optimum angle, which gives a $\pi/2$ -phase retardation, the diamond crystal works as a quarter-wave plate and it converts the linearly polarized XFEL beam generated from SACLA into circular polarization.

We made a dedicated vacuum chamber for the diamond phase retarder, shown in Fig. 2. The chamber was installed in the optics hutch at beamline **BL3** of SACLA. A diamond crystal is placed at the center of the chamber and can be rotated using the ω -20 goniometer. Three diamond crystals with different thicknesses (0.1, 0.5, and 1.5 mm) are mounted on a

crystal holder and can be selected without breaking the vacuum. These crystal thicknesses were designed to cover an X-ray energy range from 5 to 16 keV. The chamber is a standard optical component of the beamline, which is installed permanently and available for routine use in XFEL experiments that require variable polarization states.

In an X-ray polarization control experiment, a (100)-oriented diamond crystal of 1.5 mm thickness was used in the transmission Laue geometry of the 220 reflection. We monitored the X-ray magnetic circular dichroism (XMCD) signals from a ferromagnetic sample to determine the degree of circular polarization. XMCD is defined by the difference in the X-ray absorption of a sample between right- and left-handed circular polarization. The intensity of XMCD is proportional to the degree of circular polarization (P_c). We used a powder sample of a CoPt₃ alloy, which was magnetized by an electromagnet. An XFEL beam transmitted through the diamond phase retarder was introduced to the sample at an X-ray energy of the Pt L_3 edge, 11.562 keV.

Figure 3 shows the measured variation of the XMCD signal as a function of the angle of the diamond crystal ($\Delta\theta$). The origin of the horizontal axis corresponds to the exact Bragg angle for the 220 reflection. For comparison, the solid line denotes the $P_{\rm C}$ values calculated by dynamical theory. Left-handed circular polarization with a nearly perfect degree of $P_{\rm C} = -0.97$ was obtained at $\Delta\theta = 31$ arcsec. The helicity was reversed at a negative offset angle, and right-handed polarization was generated at $\Delta\theta = -31$ arcsec, but the degree of circular polarization was only $P_{\rm C} = 0.82$.

The measured variation of the polarization state was in good agreement with the calculation for a positive angular range. $P_{\rm C}$ for a negative offset was









Fig. 2. Vacuum chamber dedicated to the diamond phase retarder, installed at BL3 of SACLA.

lower than the values expected from the calculation. The purity of the circular polarization was probably degraded by the glitch observed at $\Delta\theta = -280$ arcsec (not shown). The $P_{\rm C}$ for a negative offset can be optimized by changing the azimuth of the diamond crystal by $1-2^{\circ}$ to remove the glitch. This result confirms that the helicity of circular polarization can be reversed by only a small change in the crystal angle (62 arcsec). Our system has a piezo-driven stage to quickly adjust the crystal angle, enabling angular oscillation at 100 Hz for sinusoidal and 40 Hz for rectangular motions. Helicity switching at these frequencies is feasible, enabling shot-to-shot polarization switching synchronizing with the XFEL pulses from SACLA.

In this study, we demonstrated that the diamond phase retarder works effectively for an XFEL beam to control the X-ray polarization states. Circularly polarized XFEL radiation with variable helicities has been successfully generated. The degree of circular polarization was sufficiently high for most polarization-dependent experiments. Our system has advantages in compactness, flexibility, and ability of fast polarization switching over the scheme of XFEL polarization control using specially-designed undulators. With the device currently installed, SACLA has become the only facility that can provide circularly polarized XFEL beams. This extended capability of SACLA will likely produce a number of leading-edge studies particularly in magnetic and chiral materials.



Fig. 3. Degrees of circular polarization ($P_{\rm C}$) measured as a function of the angle of diamond crystal ($\Delta \theta$).

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