Polarization control with an X-ray phase retarder for high-time-resolution pump–probe experiments at SACLA1

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1Control of the polarization of an X-ray free-electron laser (XFEL) has been performed using an X-ray phase retarder (XPR) in combination with an arrival timing diagnostic on BL3 of the SPring-8 Angstrom Compact free-electron LAser (SACLA). To combine with the timing diagnostic, a pink beam was incident on the XPR crystal and then monochromated in the vicinity of samples. A high degree of circular polarization of \(\frac{97}{\pi}\) was obtained experimentally at 11.567 keV, which agreed with calculations based on the dynamical theory of X-ray diffraction. This system enables pump–probe experiments to be operated using circular polarization with a time resolution of 40 fs to investigate ultrafast magnetic phenomena.

1. Introduction

X-ray free-electron lasers (XFELs) based on the self-amplified spontaneous emission (SASE) principle (Kondratenko & Saldin, 1980; Bonifacio et al., 1984) produce brilliant femtosecond light pulses with ultrahigh coherence in the X-ray region (Emma et al., 2010). The SPring-8 Angstrom Compact free-electron LAser (SACLA), constructed in Harima, Japan, is an XFEL facility with three beamlines: BL2 and BL3 for hard XFEL beams (Ishikawa et al., 2012; Yabashi et al., 2015), and BL1 for a soft XFEL beam (Owada, Togawa et al., 2018). Their pulse durations were evaluated to be 7.7 fs on BL3 (Inubushi et al., 2012, 2017) and below 100 fs on BL1 (Kubota et al., 2019; Owada et al., 2019). Using arrival timing diagnostic systems between the XFEL and synchronized optical laser pulses, the time resolution in pump–probe experiments has been improved to less than 100 fs (Katayama et al., 2016; Owada, Nakajima et al., 2018; Owada et al., 2019).

Polarization is a fundamental property of light. In the X-ray region, anisotropic and magnetic properties of matter have been widely investigated using various polarization states. A diamond X-ray phase retarder (XPR) (Hirano et al., 1991; Giles et al., 1994; Lang & Srajer, 1995) has been installed on BL3 of SACLA to control the polarization of the hard XFEL beam (Suzuki et al., 2014) so as to study the ultrafast dynamics of chemical bonding and magnetic states. In particular, pump–probe X-ray magnetic circular dichroism (XMCD) spectroscopy is a powerful method to investigate spin dynamics such as a demagnetization and magnetization reversal observed on...
a picosecond to femtosecond time scale (Kirilyuk et al., 2010; Takubo et al., 2017). Since the diamond XPR crystal has its diffraction plane rotated by 45° from the horizontal, the horizontally polarized incident XFEL beam is decomposed into equal parts σ- and π-polarized radiation relative to the diffraction plane. According to the dynamical theory of X-ray diffraction (Batterman & Cole, 1964), the XPR crystal produces a phase retardation between these two components near the Bragg condition, and consequently one is able to control the polarization states. In a conventional configuration [which we call a ‘mono-XPR’ (or scheme A) configuration], a monochromatic beam with a bandwidth of ΔE/E ≃ 1 × 10⁻⁴ produced with a perfect crystal monochromator is employed as the incident beam on the XPR crystal, because an energy bandwidth and/or angular divergence can cause degradation of the polarization states converted with the XPR crystal (Suzuki et al., 2014). However, one may consider locating the XPR crystal upstream of the monochromator [i.e. an ‘XPR-mono’ (or scheme B) configuration] so as to expand the applicable range of experiments with various polarization states. For example, scheme B enables us to combine the XPR system with the arrival timing monitor (TM) on BL3 of SACLA, because the TM system requires the transport of a pink beam with a bandwidth of ΔE/E ≃ 5 × 10⁻³ through the beamline optics (including the XPR crystal) located in the optics hutch (OH) (Katayama et al., 2016).

In this study, we evaluated the degree of circular polarization (P_c) of the XFEL beam in scheme B that combines polarization control with the TM on BL3 of SACLA. We experimentally confirmed a high degree of P_c, which is consistent with the calculated value and almost the same as that for scheme A (Suzuki et al., 2014).

2. Experimental

Fig. 1(a) shows the experimental setup with the XPR-mono (scheme B) configuration on BL3 of SACLA (Tono et al., 2013). In the OH, the pink XFEL beam reflected with mirrors was incident on the XPR crystal (Suzuki et al., 2014). In this study, a diamond (100) crystal 1.5 mm thick was used in a 220 symmetric Laue geometry. A branch beam of the XFEL, separated with a grating in the OH, was used for the arrival TM in experimental hutch EH1 (Katayama et al., 2016). After passing through the XPR crystal, the main XFEL beam was monochromated at the Pt L₃ edge with a pair of channel-cut crystals (CCs) in a (+, −, −, +) geometry, which maintains the height of the beam, with Si(111) reflections installed in EH1.

For comparison, we set up the conventional mono-XPR (scheme A) configuration as shown in Fig. 1(b). A double-crystal monochromator (DCM) with Si(111) reflections was used to produce the monochromatic beam. The monochromatic XFEL pulse was incident on the XPR crystal. The grating, TM and CCs monochromator were not used in this configuration.

In EH2, we installed two Kapton scattering beam monitors (BMs) to measure the intensities of the horizontal and vertical linear polarized components as shown in Fig. 1. An FePtPd film (50 nm thick) grown on an MgO(100) substrate by magnetron sputtering at 773 K was used to evaluate the P_c of the XFEL by measuring the XMCD. The sample forms an L₁₀ structure with an out-of-plane easy magnetization direction (Seki et al., 2011; Takubo et al., 2017). Using an Nd–Fe–B permanent magnet, an external magnetic field of μ₀H = ±0.59 T was applied along the surface normal of the sample film to saturate the magnetization. The sample was maintained at room temperature during measurements. We used two pairs of the sample and the permanent magnet, with opposite field directions for each pair, to measure the XMCD signal. The two samples were cut from the same parent film, and the magnetic field strengths were prepared to be nearly the same for the two configurations. The remaining systematic errors in the magnetic asymmetry were corrected. The XFEL beam was incident on the sample in the surface normal direction. To measure the absorbed intensity at the Pt L₃ edge, we detected the Pt L₃ (9.443 keV) fluorescence with a multiport charge-coupled device (MPCCD) detector (Kameshima et al., 2014), as shown in Fig. 1.

3. Results and discussion

Fig. 2(a) shows the intensities of the horizontal (Iₓ) and vertical (Iᵧ) polarization components as a function of the offset angle of the XPR crystal from the 220 symmetric Laue geometry at 11.567 keV detected with the Kapton scattering BMS in scheme B. This photon energy was selected so as to maximize the value of the XMCD. From the measured values of Iₓ and Iᵧ, the degree of linear polarization, P_L, was determined using the equation

\[ P_L = \frac{1}{Q} \frac{I_x - I_y}{I_x + I_y}, \]

where Q is the correction factor of the BMs used in this experiment. The factor was estimated to be Q =
The green and black solid curves represent the calculated values for (scheme A) and XPR-mono (scheme B) configurations, respectively. Circles and blue open circles represent the values in the mono-XPR edge as a function of the offset angle of the XPR crystal. The red solid circles and blue open circles represent the values of $P_{\text{11.567 keV}}$ for the XPR crystal angle dependence of $P_L$. At the points of $P_L = 0$, the XPR generates $\pm \pi/2$ phase retardation, and we obtain circular polarization with right- and left-helicity, respectively. A small discrepancy between the two configurations is due to the difference between the bandwidths obtained with the CCs and DCM (Suzuki et al., 2014). The monochromatic beam generated with the CCs has a bandwidth about 0.8 times smaller than that obtained with the DCM due to the difference in their reflection geometries: $(+, -, -, +)$ for the CCs and $(+, -)$ for the DCM.

Next, we estimated $P_c$ in scheme B by measuring the XMCD of the FePtPd film at the Pt $L_{3}$ edge. A magnetic asymmetry ratio, $R$, was determined by the equation

$$R = \frac{I_+ - I_-}{I_+ + I_-},$$

where $I_+$ $(I_-)$ is the absorbed intensity in the FePtPd film detected with the MPCCD for a magnetic field applied in the direction antiparallel (parallel) to the X-ray incident direction.

$I_0 = I_0'$ is the intensity of the incident XFEL beam, which is the sum of the output of the BMs. Fig. 3 shows the values of $R$ as a function of the offset angle of the XPR crystal in schemes A and B, with the calculation curves obtained by the same method as that used in a previous study (Suzuki et al., 2014). This result indicates that $P_c$ in scheme B is almost the same as that of scheme A with a high value of 0.97, which was obtained in the previous study (Suzuki et al., 2014). The uncertainty in $P_c$ in scheme B was estimated to be $\pm 0.06$. The discrepancy in the offset angles between $-20$ and $20$ arcsec is due to the influence of the bandwidth, which was seen in $P_L$ (Suzuki et al., 2014). For negative offset angles, $R$ obtained in scheme B is lower than that in scheme A, which is probably due to a glitch as discussed in the previous study (Suzuki et al., 2014).

From the experiments, we confirmed that a high degree of $P_c$ was retained in scheme B at 11.567 keV. However, according to the dynamical theory of X-ray diffraction (Batterman & Cole, 1964), a CCs monochromator could introduce a phase difference ($\delta$) between the $\sigma$- and $\pi$-polarization components, which leads to degradation of the polarization states produced with the XPR crystal. To investigate the effect of CCs on $P_c$ over a wide photon energy range, we calculated the values of $P_c$ based on the dynamical theory of diffraction. Since the angular divergence of the XFEL beam ($\sim 2\mu$rad) is sufficiently smaller than the Darwin width of the Si$(111)$ reflection $(\sim 20 \mu$rad), we only considered the effect of the energy spread. Fig. 4(a) shows rocking curves for the $\sigma$- and $\pi$-polarization components and the resulting $\delta$ and $P_c$ after the four-fold reflections of the CCs at $E_0 = 11.567$ keV. For the calculation of $P_c$, we assumed that the incident XFEL beam has circular polarization with $P_c = 1$. Although $P_c$ decreased at the edges of the rocking curves, the

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weighted average value was maintained at 0.96 at 11.567 keV. This result is consistent with our experimental results, showing that $P_C$ in scheme B has almost the same value as that in scheme A. Note that the experimental value of $P_C \approx 0.97$ in scheme B included the effects of not only the degradation by the CCs but also the narrower bandwidth than that obtained by the DCM. Fig. 4(c) shows the weight-averaged values of $P_C$ after the four-fold reflections of the CCs as a function of photon energy. This result shows that a high $P_C$ can be maintained in a high photon energy region (larger than \~10 keV), which includes XMCD for the L edges of 5d elements such as Pt, Ir and Os (Wienke et al., 1991; Schütz et al., 1989). On the other hand, the value of $P_C$ decreases drastically in scheme B in a lower energy region of less than \~10 keV, which includes the K edges of 3d transition metals and the L edges of rare earth elements (Schütz et al., 1987, 1989; Giles et al., 1994). For example, Fig. 4(b) shows rocking curves with $\delta$ and the values of $P_C$ after the four-fold reflections of the CCs at $E_0 = 7$ keV. To improve $P_C$ in this energy region, a single CC monochromator can be used in scheme B. Alternatively, a high-intensity monochromatic beam generated with the self-seeded XFEL scheme (Ammann et al., 2012; Lindberg & Shvyd’ko, 2012; Inoue et al., 2019) is available for ultrafast magnetic measurements.

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