Double phase-plate setup for chromatic aberration compensation for resonant x-ray diffraction experiments

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Abstract. An x-ray phase plate that can generate arbitrary states of linearly polarized x-rays is an indispensable device in recent resonant x-ray diffraction experiments. A thick phase plate, which is necessary to produce a high degree of linear polarization, however, considerably reduces incident x-ray intensity, particularly for low energy x-rays. Degradation of linear polarization chiefly arises from the finite energy width of incident x-rays in recent synchrotron sources, and hence we have equipped a double phase-plate setup for chromatic aberration compensation using two thin phase plates. The total linear polarization 90.5% for a single phase plate of 0.5 mm thickness was remarkably improved to 96.8% by using two phase plates of about 0.3 mm thickness at 6.1236 keV; a high degree of linear polarization was achieved without additional loss of intensity.

1. Introduction

Resonant x-ray diffraction provides a unique opportunity to examine long-range ordered structures of charge, magnetic and orbital degrees of freedom, particularly in lanthanoid and actinoid compounds. Sensitivity to higher rank multipole moments than dipoles enables the nature of hidden phase transitions to be uncovered, such as octupole order[1]. An essential aspect of this technique is polarization dependence. The symmetry of an order parameter is explored by changing the relative orientation between the order parameter and the polarization direction of x-rays. This is normally performed by an azimuthal scan, in which a sample is rotated about the scattering vector. However, during azimuthal scans, multiple scattering frequently contaminates weak resonance signals when a forbidden reflection is observed, and application to extreme conditions is difficult, for instance, when using a cryomagnet or a high-pressure anvil cell. Therefore a new and powerful method, in which polarization dependence is measured by rotating the linear polarization of incident x-rays, has recently been developed[2]. An x-ray phase plate can be used to rotate the angle of incident linear polarization. However, a thick phase plate, which produces a high degree of linear polarization, simultaneously attenuates the incident x-ray intensity. A diamond phase plate of 1.0 mm in effective thickness transmits only 2% of incident x-rays at 6 keV. This is not a minute problem for resonant x-ray diffraction experiments at low x-ray energies. In this paper, we describe a double phase-plate setup for chromatic aberration compensation recently installed at SPring-8. This device achieves both a
high degree of linear polarization and low attenuation and is suited to resonant x-ray diffraction experiments.

Before proceeding to the experimental section, we briefly explain an x-ray phase plate. An x-ray phase plate utilizes the birefringence near a Bragg condition and is usually a perfect single crystal of Si or diamond. Linear horizontal polarization of incident x-rays is transformed to an arbitrary polarization state by using a phase plate, which produces a phase shift $\delta$ between the components of the electric field perpendicular ($\sigma$) and parallel ($\pi$) to the diffraction plane[3]. If the two perpendicular components have equal amplitude, circularly polarized x-rays are generated for a $\pi/2$ phase shift, and linear vertical polarization is achieved for a $\pi$ phase shift. In the same way, for a $\pi$ phase shift, one can rotate the linear polarization state in any direction by changing the relative amplitude of the two components.

Currently the most popular geometry of an x-ray phase plate uses the forward diffracted beam away from the Bragg angle. For that geometry, dynamical theory tells that to a good approximation the phase shift $\delta$ between the two components of the transmitted x-rays is given by

$$
\delta = -\frac{\pi}{2} \left[ \frac{r_e^2 \text{Re}(F_h F_{\bar{h}})}{\pi^2 V^2} \frac{\lambda^3 \sin(2\theta_B)}{\theta - \theta_B} \right] t = -\frac{\pi}{2} \frac{A}{\Delta \theta} \, ,
$$

where $r_e$ is the classical electron radius, $F_h$ and $F_{\bar{h}}$ the structure factors of the $hkl$ and $\bar{h}\bar{k}\bar{l}$ reflections, respectively, $\lambda$ the wave length of x-rays, $\theta_B$ the Bragg angle, $V$ the volume of the unit cell, $\theta$ the glancing angle on the $hkl$ plane, and $t$ the x-ray path length in the crystal. In short, the phase shift $\delta$ is inversely proportional to the angular offset $\Delta \theta$ and thus is controlled by varying $\Delta \theta$.

2. Experimental

Experiments were carried out at beamline BL22XU in SPring-8. Two setups were employed. In one setup, a single diamond phase plate of 0.5 mm thickness was used, and in the other setup, two diamond phase plates of 0.3 mm thickness were used. All the diamond phase plates had a [100] surface, and the (111) reflection in asymmetric Laue geometry was used. Each diamond crystal was mounted on a rotation stage to adjust $\theta$ and was further mounted on a large circle goniometer. There were four mount points located in the first, second, third and fourth quadrants and the angle between neighboring mount points was 90° as shown in Fig. 1.

In the experiment with two phase plates, the first phase plate and the second phase plate were placed in the first quadrant and the second quadrant, respectively. The rotation angle $\phi_{pr}$ about

![Figure 1](image_url)

**Figure 1.** Schematic illustration of the experimental setup of two phase plates. A view from the upstream side. Two phase plates, each of which is mounted on a goniometer head and a rotation stage ($\theta_1$ or $\theta_2$), are also mounted on a large circle goniometer ($\phi_{pr}$). Rotating $\phi_{pr}$ rotates both phase plates about the x-ray beam. In this figure, the first phase plate and the second phase plate are placed in the first quadrant and the second quadrant, respectively, and the rotation angle $\phi_{pr}$ is equal to $-45^\circ$. 
the incident x-ray beam modifies the relative amplitude of the two components and thus is essential for producing an arbitrary linear polarization. The beamsize at the phase plate(s) was 0.5 mm (horizontal) × 0.7 mm (vertical). For the single phase-plate setup, the phase plate works as a half-wave plate. In contrast, for the two phase-plate setup, each phase plate works as a quarter-wave plate and the total phase shift becomes $\pi$.

The polarization state of the transmitted x-rays was analyzed with a standard x-ray polarization analyzer. We used the Al (220) reflection and tuned the photon energy to 6.1236 keV so that the scattering angle was equal to $90^\circ$. The polarization analyzer was rotated about the transmitted x-ray beam by an angle $\phi_A$ and at each point the integrated intensity was obtained by rocking the analyzer crystal ($\theta_A$). The obtained integrated intensities were then fitted to

$$I = I_0 \frac{1}{2} (1 + P_3 \cos 2\phi_A + P_1 \sin 2\phi_A),$$

(2)

and the Stokes parameters $P_3$ and $P_1$ were determined. The degrees of linear polarization were finally estimated from the quantity $\sqrt{P_3^2 + P_1^2}$.

3. Results and Discussion

Here we start by briefly describing the principles of off-axis and chromatic aberration compensation for phase plates (Fig. 2)[4, 5]. Solid black curves indicate $\delta \propto \Delta \theta^{-1}$ (eq. 1), and the width of the gray vertical lines represents the finite angular divergence of incident x-rays. The energy width of incident x-rays are depicted by dotted blue and broken red curves for higher and lower energies, respectively. The first phase plate, which is located in the first quadrant (bottom left in the figure), is initially irradiated with incident x-rays (a gray line). Because of the finite angular divergence of incident x-rays, even for monochromatic x-rays (a solid black curve), the resultant phase shift also has finite width. Similarly, since the finite energy width of the x-rays shifts the Bragg angle to the right and left, the solid black curves also shift to dotted blue and broken red curves. Accordingly the energy width gives rise to a finite spread in phase shift as well. For the second phase plate, which is placed in the third quadrant (top right), the definition of $\theta$ is reversed. Hence although x-rays at the left edge of the gray vertical line gain a greater phase shift than the average at the first phase plate, they lose the excess shift at the second phase plate. Likewise, a small phase shift at the first phase plate for x-rays at the right edge is recovered at the second phase plate. This is the principle of off-axis aberration compensation. In the second and fourth quadrants, the roles of $\pi$ and $\sigma$ polarizations are reversed. By definition, this changes the sign of $A$ in eq. 1. Hence, the first quadrant is transformed into the fourth quadrant, and the third quadrant is transformed into

Figure 2. Illustration showing how two phase plates compensate for off-axis and chromatic aberrations. The abscissa is the angular offset from the Bragg angle $\Delta \theta$ and the ordinate is the phase shift $\delta$. Solid black curves indicate $\delta \propto \Delta \theta^{-1}$ (eq. 1) for x-rays of central energy and dotted blue and broken red curves correspond to x-rays with higher and lower energies, respectively. The width of the vertical gray lines indicates finite angular divergence of incident x-rays. See text for details.
Rotation about the incident x-rays $\phi_{pr}$ (degrees)

(a) $\phi_{pr}=45^\circ$

(b) $\phi_{pr}=45^\circ$, $\phi_{pr}=60^\circ$, $\phi_{pr}=75^\circ$, $\phi_{pr}=90^\circ$

Figure 3. Total linear polarization $\sqrt{P_3^2 + P_1^2}$ measured as a function of $\phi_{pr}$ for (a) a single phase plate and (b) two phase plates with chromatic aberration compensation (the first phase plate at the first quadrant and the second phase plate at the second quadrant). The degree of linear polarization is low at $\phi_{pr} = \pm 45^\circ$ (vertical linear polarization). The intensity integrated over $\theta_A$ is also plotted as a function of $\phi_{pr}$ in polar coordinates for several $\phi_{pr}$s. Note that the square-root of the intensity $I^{1/2}$ is plotted instead of the intensity $I$ itself in order to enlarge details. A marked improvement in polarization from the single phase plate is visible at $\phi_{pr} = 45^\circ$ and rotation of linear polarization is clearly displayed.

Experimentally, the effect of the second phase plate was apparent. The total linear polarization $(P_3^2 + P_1^2)^{1/2}$ for the single phase plate of 0.5 mm thickness is shown in Fig. 3(a) as a function of $\phi_{pr}$. The degree of linear polarization was low at $\phi_{pr} = \pm 45^\circ$ and was at most 90.5%. On the other hand, in the experiment of two phase plates of 0.3 mm thickness (Fig. 3(b)), the total linear polarization was considerably improved, particularly at $\phi_{pr} = \pm 45^\circ$ and reached to 96.8%. Polarized states are also recognized in the inset graphs, where in order to enlarge details, the square-root of the intensity $I^{1/2}$ integrated over $\theta_A$ is plotted as a function of $\phi_{A}$ in polar coordinates. Clearly, the polarization state at $\phi_{pr} = 45^\circ$ in Fig. 3(b) is much purified compared with that in Fig. 3(a). Rotation of the angle of linear polarization with increasing $\phi_{pr}$ is also well demonstrated.

The experimental results were also supported by calculation. Total linear polarization at $\phi_{pr} = 45^\circ$ was calculated for single phase plates of 0.5 mm and 1.2 mm thicknesses and for two

|                   | 1st(0.5mm) | 1st+2nd | 1st+3rd | 1st+4th | 1st(1.2mm) |
|-------------------|------------|---------|---------|---------|------------|
| $(P_3^2 + P_1^2)^{1/2}$ | 0.901      | 0.986   | 0.945   | 0.997   | 0.984      |

Table 1. Total linear polarization calculated at $\phi_{pr} = 45^\circ$ for single phase plates of 0.5 mm thickness and 1.2 mm thickness and two phase plates of 0.3 mm thickness. For two phase plates, calculations were performed under three configurations. See text for details.
phase plates of 0.3 mm thickness, and the calculation results are indicated in Table 1. For two phase plates, calculations were performed under three configurations, namely, combinations of the first and second quadrants, the first and third quadrants, and the first and fourth quadrants. We set the coefficient $A$ to be $0.0176 \degree^{-1}$ for a 0.3 mm phase plate. We convoluted the phase shift $\delta$ using Gaussian profiles of $0.002^\circ$ and $0.8$ eV (full width at half maximum) for the angular divergence and the energy width, respectively, of the incident x-rays. The horizontal focusing mirror gives rise to the angular divergence $0.002^\circ$, and we assumed the same vertical angular divergence for simplicity. In addition to quantitative agreement with experiments, it was found that the chromatic aberration compensation is very effective for obtaining a high degree of linear polarization. A calculation also shows that a single phase plate of 1.2 mm thickness generates a linear polarization comparable to 98.6%. It is hence interesting to compare the transmission rate of two 0.3 mm phase plates (total effective thickness 0.603 mm) with that of a single phase plate of 1.2 mm thickness (effective thickness 1.206 mm) for x-rays of 6.1236 keV. Although the both setups generate the same degree of linear polarization, the transmission rates of the former and the latter are 12% and 1.4%, respectively. Therefore we have produced a high degree of linear polarization without significant loss of intensity.

To summarize, we have equipped a double phase-plate setup for chromatic aberration compensation. The use of two thin phase plates realized both a high degree of linear polarization and an acceptable range of reduction in incident x-ray intensity adequate for resonant x-ray diffraction experiments. Finally, we note that similar developments were performed at ESRF[5] and DESY[6].

Acknowledgments
This work was partly supported by Grants-in-Aid for Scientific Research on the Basic Research A (No. 21244056) and B (No.24340087) from the Ministry of Education, Culture, Sports, Science and Technology, Japan. The synchrotron radiation experiment was performed under Proposal No. 2010A3711 at BL22XU in SPring-8.

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