X-ray Magnetic Absorption and Scattering (XAFS)

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1.Introduction

In 3rd generation synchrotron radiation (SR), one of the most promising items may be control and characterization of polarization state. In particular, they are a crucial factor to study magnetic properties using the various techniques inscattering, absorption, diffraction and emission experiments. Indeed, low emittance SR such as Spring-8 can efficiently realize this performance, and for this reason a combination of linear undulator and phase-retarder was decided on the design of beamline 39XU.

Experimental test of instruments for X-ray magnetic absorption and scattering (XMAS) has successfully demonstrated that the apparatus has the feasibility for studying magnetism. In this report we present the results of X-ray resonance magnetic scattering X-ray magnetic circulardichroism,X-ray magnetic dillraction, and ATS scattering, in which the phase retarder played an essential role for the control of both linear and circular polarization states. Finally, we view some studies to be extended on the beamline in near future.

2.Experimental Utility

Linearly polarized X-ray is led to the experimental hutch after monochromatization by the standard double-crystal monochromator and rejection of higher harmonics by the mirror. The apparatus for XMAS is composed of a phase retarder assembly, a 3-circle diffractometer, and another 4-circle goniometer for polarization analysis, as schematically shown in Fig.1.

The phase retarder, placed at the most upstream in the hutch, perfectly functioned both to convert linear polarization to circular one (as a $\lambda/4$-phase plate) and to generate vertical linear polarization (as a $\lambda/2$-phase plate). A synthetic diamond (111) crystal slab 0.73 mm in thickness was fixed on the crystal holder of a standard goniometer, tilted by 45 ° from the polarization plane of the incident X-ray, and was operated around the 220 reflection in transmission Laue geometry.

The 3-circle diffractometer, installed on a large *x*-*z* translation stage, consists of a 2θ-arm, an ω-circle to carry an electromagnet and a cryostat, and an additional ω-arm, which will be available for a fluorescence detector or an optional sample orientation. An electromagnet, designed to supply the magnetic field up to 2 Tesla, and a He-gas closed cycle refrigerator cooling down to 10 K will be installed,

Another 4-circle goniometer mounted on the 2θ-arm of the diffractometer functions as a polarization analyzer assembly. The scattered X-rays are separated into the π and σ polarization components to specify the scattering matrix. For the experiments in the photon energy range of 7keV, aSi(331) channel-cut crystal was prepared and mounted on the χ-circle together with a scintillation counter. Degree of linear or circular polarization was estimated from the angle dependence of X-ray intensity.

The detector used in this work includes a Si(Li)SSD, scintillators, and ionization chambers. Although Lab VIEW was mainly used as software for the present experiments, SPEC running on LINUX system will be installed.

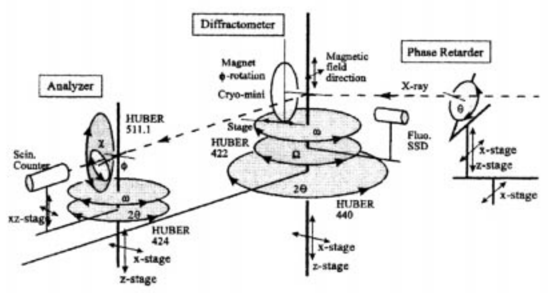

Fig.1 Schematic view of the XMAS apparatus.

3. Experimental Tests

At an interesting absorption edge the phaseplate was adjusted to an appropriate angular condition and the degree of polarization was estimated, then an experiment using the linearly or circularly polarized X-rays was carride out under the condition.

3.1 Characterization of polarization

The dependence of X-ray intensity on the angle of χ-circle is expressed as follows:

$I(\chi)=S_0+S_1\cos 2\chi+S_2\sin 2\chi$, (1)

where S_0, S_1, and S_2 represent the components of Stokes parameter, χ is the azimuthal angle of the 4-circle goniometer. Degree of polarization can be derived from these parameters.

Since this beamline was equipped with the standard linear undulator,the linearly polarized X-rays (π polarization) are originally available. Degree of linear polarization P_L of the incident X-ray monochromatized at 7.1195 keV was evaluated to be 0.998 by a logarithmic fitting to eq.(1). Such a high rate is suitable for not only the magnetic scattering but also a conversion to other polarization states by the phase retarder.

Indeed, the linearly polarized incident X-ray was converted to the circularly polarized X-ray using the diamond phase-plate. We adjusted an offset angle from the Bragg condition so as to produce a $\pi/4$phase shift between the π and σpolarizations. Degree of circular polarization Pc was also estimated to be 0.99. Since the offset angle depends on wavelength, Bragg angle, X-ray path length in the crystal, etc.,the angular conditions were previously determined for XMCD measurements at the Fe and Co *k*-edge and the Pt L_3-edge.

Another conversion is to generate a vertical linear polarization (σ polarization) as a $\lambda/2$-phase plate. Degree of linear polarization P_L was also estimated to be 0.82. A deterioration of P_L is possibly due to the contamination of simultaneous reflections from the phase-plate and/or higher harmonics of the undulator light. The angle dependence of X-ray intensity, represented using the polar coordinate, is illustrated in Fig.2 for each of the polarization state.

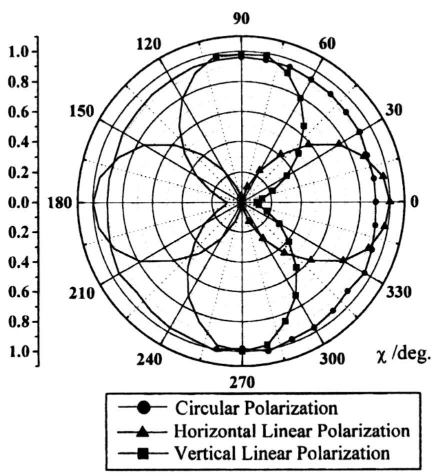

Fig. 2 Angle dependence of X-ray intensity.

3.2 Linear polarization

Using the phase-plate, we can easily alternate between the π and σ polarizations without rotating any axis of sample. This performance was tested by the following diffractometries.

3.2.1 X-ray resonance magnetic scattering (XRMS)

To estimate an efficiency of the diffractometer, we have measured Fe K-edge XRMS of the 200 Bragg reflection in pure Fe. A disk 6 mm in diameter (100) oriented Fe single crystal was used. We paid attention to observation of a difference between dichroic spectra using the π and σ polarizations.

The magnetic effect is manifested by the flipping ratio, defined as

$$R_a = (I^{\uparrow} - I^{\downarrow}) / (I^{\uparrow} + I^{\downarrow}), \quad (2)$$

where $I^{\uparrow}$ ($I^{\downarrow}$) indicates the intensity for the magnetization parallel (antiparallel) to the cross products ($\mathbf{k}\times\mathbf{k}'$) of the wave vectors of incident and scattered X-rays. The magnetic field of 2 kOe was applied to the (100) plane.

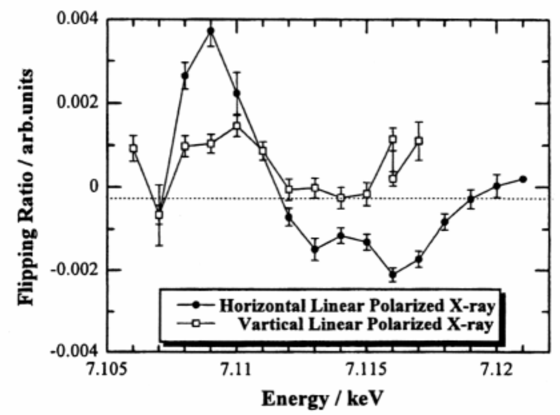

Fig.3 Flipping ratios at the Fe K-edge.

Figure 3 shows the R_a spectra at the Fe *k*-edge using the horizontal (π) or vertical (σ) linearly polarized X-ray. The usual spectrum, recorded by the π polarization, has dispersion type shape near the edge, which is in good agreement with the early data taken at 2nd generation SR facility [1]. On the other hand, the spectrum using the σ polarization indicates no change of the sign in the higher energy side. This difference originates from the polarization contributions to the cross section, and will give us rigorous information on a distinction between the dipolar and quadrupolar transitions. A combination with the polarization analysis of the scattered X-ray extends the XRMS technique into antiferromagnets.

3.2.2 Anisotropy of the tensor susceptibility (ATS) scattering

A demonstrative phenomenon for change of the polarization state is ATS scattering, wihch can be observed as forbidden reflection in the vicinity to absorption edge due to a breakdown of extinction rule. For this experiment a single crystal of FeS_2 (Pyrite) was prepared. The angle dependence I (χ) of the (001) forbidden reflection was measured around the Fe *k*-edge using the polarization analyzer assembly. As a result, a change of the π or σ polarization due to the ATS scattering is easily confirmed, that is, when the incident X-ray has the π polarization, the scattered beam is completely converted to the σ polarization, *vice versa* [2]. The σ polarization is obtainable from the phase retarder operated as a $\lambda/2$-phase plate. Efficiency of the diamond phase-plate was clearly demonstrated.

3.3 Circular polarization

Circularly polarized X-rays are indispensable for measuring magneto-optical effects. The phase plate has high performance to generate the circularly polarized X-ray with a high-rate and a tunabiolity of *Pc*.

3.3.1 X-ray magnetic circular dichroism (XMCD)

When the offset angle was adjusted to about 170 arcsec at the Fe K-edge, the rate of *Pc* > 0.99 was obtained. Helicity can be easily alternated between the offset angles of ±170 arcsec. Figure 4 shows the Fe *k*-edge XMCD in pure Fe, recorded in transmission mode while magnetic field was reversing under the fixed plus helicity. A piece of Fe-foil 5μm in thickness was inserted into the magnetic field of 1 kOe, whose direction was tilted by 60° away from the incident X-ray. This spectrum clearly shows a larger dichroic signal, a better statistical accuracy, and a better efficiency in comparison with the data taken at 2nd generation SR facility [3]. These advantages result from the high-flux beam and the high-rate of Pc. When the offset angle was changed the sign, a reversal of XMCD sign was confirmed at the Co k-edge in pure Co foil, as shown in Fig.5. At the Pt L_3-edge the offset angle was adjusted to ± 20 arcsec, and the dichroic signal reached the amplitude of 4% in 61.5 at % Pt-Fe alloy. Using this feature, we measured the dependence of XMCD intensity on the offset angle, which gives us the variation of Pc as a function of offset angle.

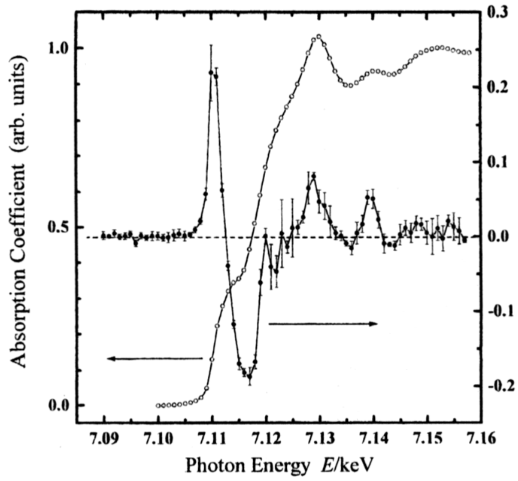

Fig.4 XMCD at the Fe *k*-edge in pure Fe.

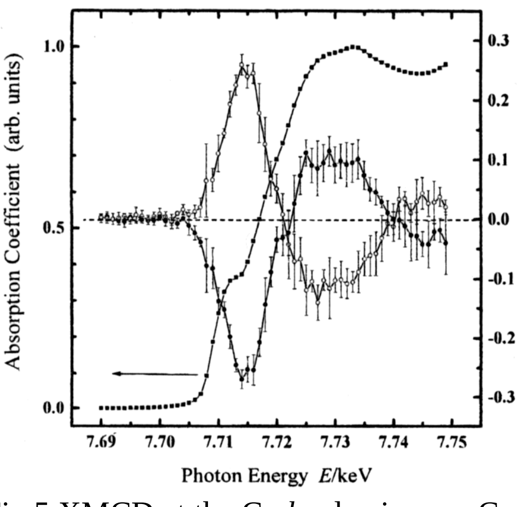

Fig.5 XMCD at the Co *k*-edge in pure Co.

These tests demonstrate that the apparatus is available to XMCD and applicable to not only the experiments under extreme conditions but also other magnetic effects, *e.g.*,dichroism in multielectron excitation, resonant X-ray emission, etc. An extension to linear dichroism or exotic materials is also possible.

3.3.2 X-ray magnetic diffraction (XMD)

X-ray magnetic diffraction is a technique to directly measure the spin and orbital moment densities using elliptically polarized X-rays. The degrees of linear and circular polarization can be easily controlled by tuning the offset angle of the phase plate, so that the XMD measurement by a monochromatic X-ray method has been applied to Fe single crystal.

The Bragg diffraction from a (220) plane of pure Fe was measured by tuning photon energy and polarization state. When we adjust the photon energy to E = 8.65 keV (λ= 1.433 Å), the configuration of 90 ° scattering is maintained. Magnetic effect is manifested by the flipping ratio described as eq. (2). In this test we observed the variation of the flipping ratio R_a with the offset angle, as shown in Fig.6 in which R_a is proportional to the polarization factor $Pc/(1-P_L)$. The ratio reached the amplitude of 1%, which was about twice as large as that measured in 2nd generation SR facility [4].

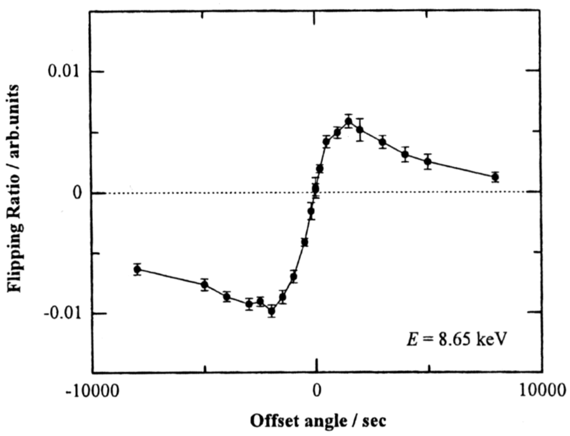

Fig.6. Flipping ratio as a function of offset angle.

4. Development in near future

A modulation of helicity will be realized by an alternation between plus and minus offset angles. This technique will open out the measurements in polarization-modulation mode, which will lead to an improvement of data accuracy and remove some experimental limitations.

References

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