2 Newly Develped Experimental Systems

2-1 Development and Applications of an Apparatus for Soft X-Ray Resonant Magnetic Scattering

An apparatus for soft X-ray resonant magnetic scattering (SXRMS) experiments with polarization-variable undulator radiation has been developed [1]. This instrument is intended for exclusive use for experiments on X-ray magnetic circular (XMCD) and linear (XMLD) dichroism in SXRMS processes in order to study the correlation and competition between electronic and magnetic states and long-range spatial magnetic structure. The apparatus consists mainly of mechanisms for high-precision sample/detector rotations, a temperature-controllable sample holder, a gap-variable permanent magnet, horizontally and vertically beam-defining slits, a main measurement chamber, a sample-preparation chamber, and a load-locked chamber. The whole system is ultrahigh-vacuum compatible and is to be fully computer-controlled. It has been installed on BL-16A for expected user experiments, as shown in Fig. 1.

The sample rotation (θ) and two-angle detector rotations (2θ, ϕ) are simultaneously or independently achieved with an accuracy of ±0.001° from outside the main chamber through bellows using stepping motors (Fig. 2). Angle θ represents the rotation within the light-incidence plane while angle ϕ stands for the rotation perpendicular to the plane. The sample can be cooled down to ~5 K using a helium-flow cryostat, with the temperature controllable over a range of ~6-300 K. The horizontal magnetic field at the sample position can be manually changed over a range of ~1-2 kG by adjusting the permanent-magnet gap from outside the vacuum chamber while maintaining the closed loop of the magnetic flux (Fig. 2). A Si photodiode is used as a detector

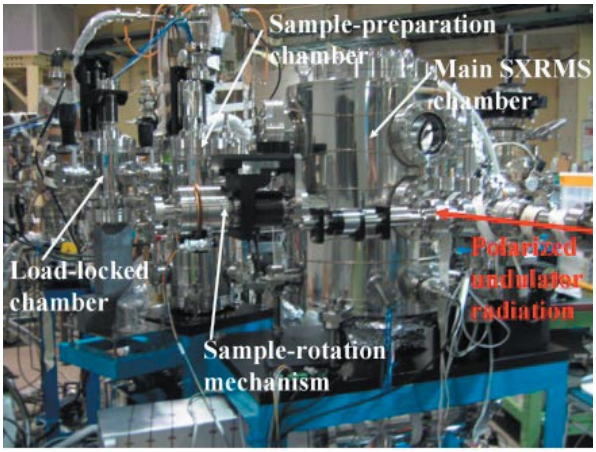

Figure 1
Upstream view of the SXRMS apparatus installed on the polarization-variable undulator beamline BL-16A. The main SXRMS chamber is seen in the foreground.

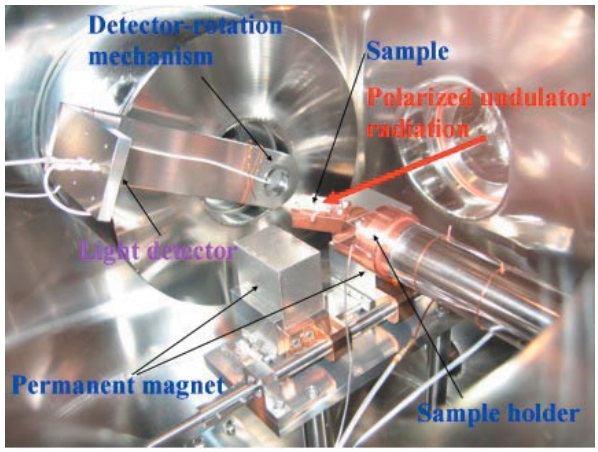

Figure 2
Main inner mechanisms for the SXRMS instrument. The magnet gap is manually adjustable from outside the chamber with the closed loop of the magnetic flux maintained.

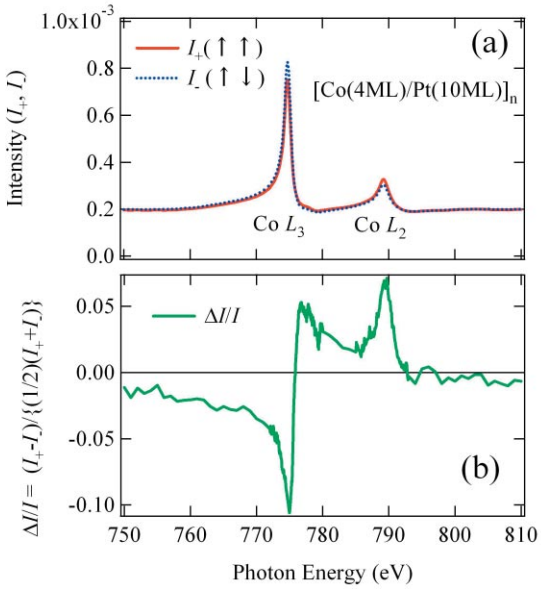

Figure 3
(a) The first-order diffraction intensities (I_+ and I_-) as functions of circularly polarized photon energy for a Co(4ML)/Pt(10ML) multilayer.
(b) XMCD spectrum $\Delta I/I = (I_+ - I_-)/\{1/2(I_+ + I_-)\}$ for the first-order diffraction peak.

with horizontal and vertical apertures placed in front of it. The preparation chamber allows *in situ* sample preparation, such as a cleavage, diamond filing, and sample evaporation. The load-locked chamber enables one to quickly exchange sample substrates. The sample can be transferred under ultrahigh vacuum.

Preliminary SXRMS experiments were made on Co(4ML)/Pt(10ML) multilayers using circularly polarized soft X-rays emitted from the APPLE II undulator at BL-16A. A magnetic field of $\boldsymbol{B}$ ~ 2 kG was applied nearly parallel to the sample surface plane. The photon energy was changed over a range covering the Co $L_{3,2}$ edges

with the grazing incidence angle kept constant. The resonantly scattered light intensity (I) was measured as a function of the scattering angle with the photon energy set at the Co L_3 edge, the result showing an SXRMS pattern with clear zeroth- and first-order diffraction peaks. The helicity-dependent scattered light intensity ($I_\pm$) was also measured as a function of photon energy (SXRMS spectra) for the zeroth- and first-order diffraction peaks with the photon helicity (h) parallel and antiparallel to -$\boldsymbol{B}$. Figure 3(a) shows the first-order SXRMS spectra, I_+ and I_-, for h =±1, respectively. Figure 3(b) shows an XMCD spectrum, $\Delta I/I = (I_+ - I_-)/\{1/2(I_+ + I_-)\}$, for the first-order superlattice diffraction peak. An XMCD signal was clearly observed at the Co $L_{3,2}$ edges in the first-order SXRMS as well as in the zeroth-order SXRMS (not shown here). This result shows a ferromagnetic alignment of the in-plane magnetized Co layers across the nonmagnetic Pt layers.

We plan to install a polarization analyzer in the main chamber to study the polarization changes on SXRMS. We also intend to apply our apparatus, equipped with a position-sensitive detector, to investigations of order-disorder transitions by observing speckle patterns with spatially coherent radiation.

REFERENCE

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2-2 Introduction of an X-Ray Phase Plate to the Wevelength-Dispersive Spectrometer at AR-NW2A

The wavelength-dispersive spectrometer is a powerful instrument for investigating the local structure and electronic state of a target element during fast reaction processes [2]. For example, catalytic chemistry research has been intensively carried out at AR-NW2A [3]. Another promising application is spin-dynamics research of magnetic materials using X-ray magnetic circular dichroism (XMCD). In order to explore this direction, we have introduced an X-ray phase plate to the wavelength-dispersive spectrometer at AR-NW2A.

Figure 4 shows the experimental setup for time-resolved XMCD experiments. The spectrometer is mainly composed of a bent-crystal polychromator, a sample stage, and a linear X-ray detector. Incident white X-rays are focused and dispersed by the bent-crystal, so that the photon energy in the focused beam varies as a function of convergence angle through the focus. The specimen is placed at the focus. By measuring the X-ray intensity distribution across the beam behind the focus, in the presence and absence of the specimen, the absorption spectra are obtained. For the XMCD experiments, we introduced an X-ray phase plate [4] between the bent-crystal and the focus to convert linear polarization

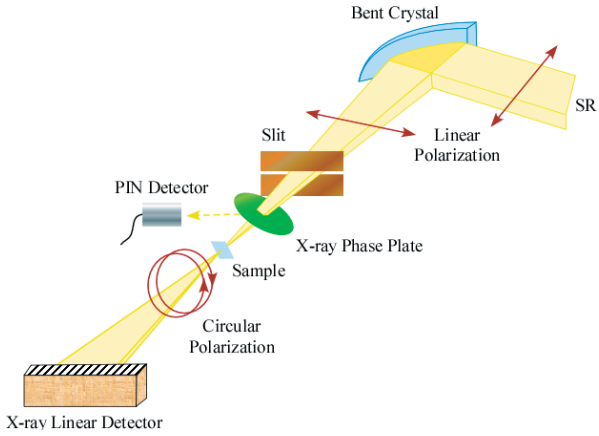

Figure 4
Experimental setup for time-resolved XMCD experiments. The optics is mainly composed of a bent-crystal polychromator, an x-ray phase plate, a sample stage, and a linear x-ray detector.

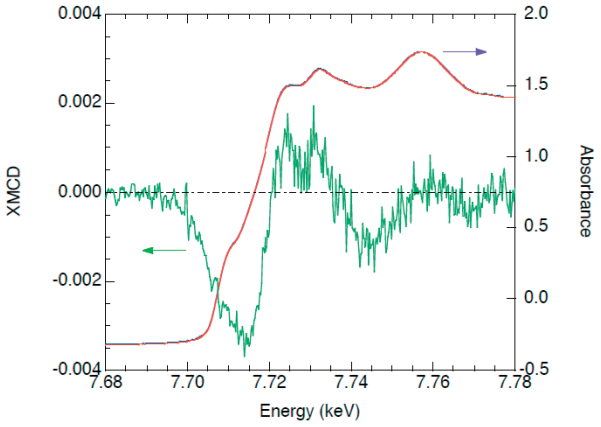

Figure 5
XMCD spectra of a 4 μm thick Co-film around the Co-K absorption edge (λ ~ 0.1608 nm).

to circular polarization. The helicity of the beam can be easily switched by slightly changing the incident angle of the phase plate. It is important to keep the degree of circular polarization (Pc) constant over the entire energy range. This matching condition is expressed as follows:

$$\Psi = \cos^{-1}\left[\frac{1}{2}\frac{\tan\theta_2}{\tan\theta_1}\left(1-\frac{q}{p}\right)\right]$$

where Ψ is the angle between the diffraction plane of the phase plate and the horizontal plane, p the distance from the source to the bent-crystal, q that from the bent-crystal to the focus, and θ_1 and θ_2 the Bragg angles of the bent crystal and the phase plate, respectively. In order to produce circular polarization, it is necessary to design the optical system so that Ψ is close to 45°.

The XMCD spectrum of a 4 μm thick Co-film was recorded around the Co-K absorption edge (λ ~ 0.1608 nm). A flat Si(111) crystal was bent using a thermostated holder (p = 30 m, q = 0.259 m, R = 2 m). A type-IIa synthetic diamond (001) crystal of 0.7 mm thickness

was used as the phase plate. The diamond crystal was adjusted in the vicinity of the Laue-case 111 diffraction condition to convert linear polarization to circular polarization. The Ψ-angle was set at 37.5°. The measurement time for a single data set was 450 msec. To improve the statistical precision of the data the measurement was repeated 10 × 8 = 80 times, leading to a total measurement time of 36 sec. The obtained XMCD spectrum is shown in Fig. 5, and it agrees well with a spectrum recorded using the energy-scanning method. This result shows that combining the wavelength-dispersive spectrometer and the X-ray phase plate opens up new possibilities for spin-dynamics research of magnetic materials.

REFERENCES

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[3] Y. Inada, A. Suzuki, Y. Niwa and M. Nomura, *AIP Conf. Proc.,* **879** (2006) 1230.
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2-3 Commissioning of Resonant Scattering Equipment for Use in the Soft X-Ray Energy Region

We have newly developed an apparatus for soft X-ray scattering in materials science research fields such as strongly-correlated electron systems (SCES) and nanomaterials. The scattering equipment has been constructed at the BL-16A undulator beamline, where fast polarization switching at a frequency of ~ 10 Hz will be available from 2010.

The energy region of 200 - 1500 eV at BL-16A covers the L absorption edges of the 3d transition metal elements, the M absorption edges of the 4f rare metal elements, and the K absorption edges of the light elements, which play an important role in probing directly electronic states related to physical properties.

Consequently, we are able to reveal the significance of the hybridization between the 3d electronic state and the oxygen electronic state in the SECS.

In addition, by using the circular and linear polarization characteristics, we will further study the magnetic properties and local symmetry of orbital states at element-selective sites.

The soft X-ray scattering equipment mainly consists of 1) a detector section, 2) a sample goniometer head, and 3) a refrigerator.

1) Three kinds of detectors are set up; an MCP (Hamamatsu), a photodiode(IRD), and a two-dimensional CCD camera (Princeton Instruments).The distance between the sample position and the detectors (2θ arm length) is approximately 130 mm, and the angle between 0° and 170° is available for measuring.

2) The beam spot size at the focal point in the vertical (horizontal) direction is 50 - 100 μm (100 - 200 μm). The centering of the sample position for the beam is adjusted by XYZ motions of the goniometer head.

3) The samples are cooled by a He-flow refrigerator. It takes less than one hour to reach low temperatures (~10 K).

The soft X-ray scattering equipment works well and efficiently.

Figure 7 shows the reflectivity for a hybridized thin film of polystyrene and polyvinylpyridine with a thickness of 600 Å. We can see a periodic oscillation of 0.005 A^{-1} for the profile, which corresponds to the domain structure.

Although the measuring time was only 100 msec for each point, the q-dependence of the reflectivity is clearly observed over a range of more than 10^6. We have also performed soft x-ray scattering on a perovskite man-

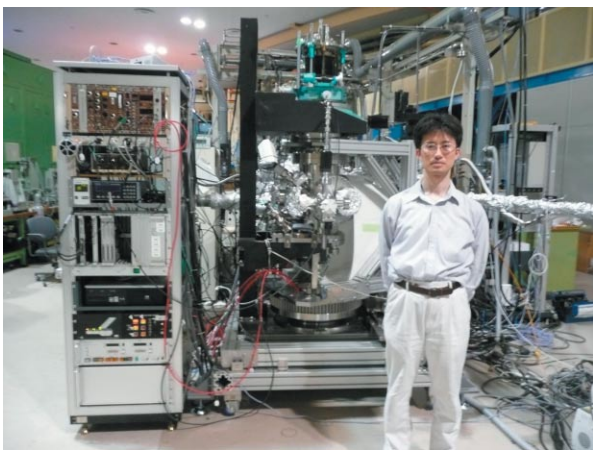

Figure 6
View of the soft X-ray scattering equipment.

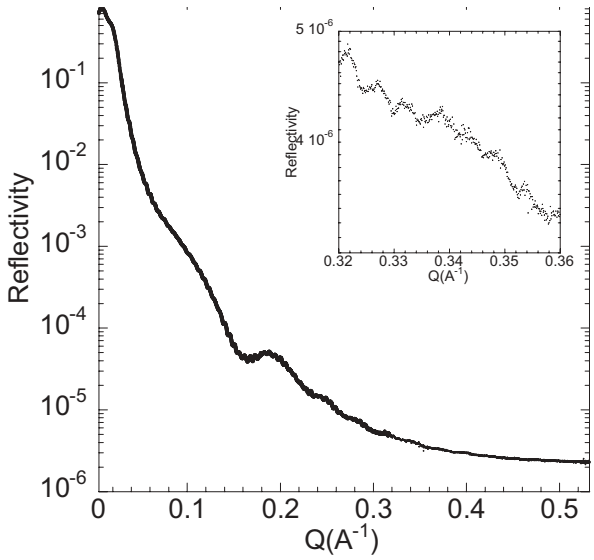

Figure 7
Reflectivity of a hybridized thin film of polystyrene and polyvinylpyridine recorded with a photon energy of 543 eV for 100 msec per point. A clear periodic oscillation is observed, as shown in the inset.

ganite (La,Sr)MnO_3/$SrTiO_3$ superlattice thin film and layered perovskite manganite $Nd_{2-x}Sr_xMnO_4$ (x = 2/3), which show orbital/charge ordered states.

In the near future, we will establish automatization of both the centering adjustment of the sample position and the measuring of the temperature dependence of satellite peak intensities. We will further add the functionality of detecting inelastic scattering signals to the present equipment.

At KEK, the Condensed Matter Research Center (CMRC) was established in 2009, where quantum beams of synchrotron radiation, neutrons, and muons will be utilized to reveal the mechanisms of interesting physical properties. We believe that a complementary utilization of soft X-ray scattering will be useful for probing electronic states in detail.

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