Development of an in-vacuum diffractometer for resonant soft X-ray scattering

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\textbf{Abstract.} Resonant soft X-ray scattering measurements become one of powerful techniques to probe charge, orbital and spin orderings in strongly correlated electron systems. We have developed new in-vacuum diffractometer with a large vacuum chamber to solve experimental limitations of in-vacuum measurement in soft X-ray energy region, and for future instrumental developments. In this paper, the features of the diffractometer are presented.

1. Introduction

Various interesting physical properties, such as colossal magnetoresistance effect and magneto-electric effect, were discovered in strongly correlated electron systems (SCES). There the strong correlation among orbital, charge, and spin degrees of freedom play important roles. The study of these electronic ordering states is essential to understand the phenomena microscopically. Resonant X-ray scattering (RXS) at the \textit{K}-edge becomes a powerful tool for observing the spatial ordering of charge and orbital degrees of freedom in \textit{3d} transition metal oxides. \cite{1, 2}

The RXS signal at \textit{K}-edge (\textit{1s} \rightarrow \textit{4p} transition energy) reflects the \textit{4p} electronic state. On the other hand, the RXS at \textit{L\textsubscript{2,3}}-edge (\textit{2p} \rightarrow \textit{3d} transition energy) can probes the \textit{3d} electronic state directly, and the signal of the resonant magnetic scattering is strongly observed at the \textit{L\textsubscript{2,3}}-edge of \textit{d} electron system. Moreover, \textit{K}-edges of \textit{O}, \textit{S}, and \textit{P} ions, which play an key role for the itinerancy through the hybridization with the metal ion, becomes also observable by using soft X-ray. Hence the resonant soft X-ray scattering (RSXS) measurements become to be performed globally. \cite{1, 2}

We have developed a new in-vacuum diffractometer to perform the RSXS experiment. The RXS signal reflecting the change of physical properties is weak in general. Therefore, the improvement of signal-to-noise ratio is important to detect the signal. Here we built the diffractometer with a large vacuum chamber, and slit and detector are installed in the vacuum chamber to reduce the background, such as fluorescence. The specifications of the diffractometer and future plans are presented.
2. Instrument

2.1. Diffractometer

We developed a general in-vacuum diffractometer dedicated to the resonant soft X-ray scattering measurement. This diffractometer with a large vacuum chamber is shown in Fig. 1. Utilizing the large vacuum chamber, we can install several devices, the slits, detectors, and so on, on the diffractometer. Moreover, it has a possibility to install new devices in future. Vacuum pumping is performed by a turbo molecular pump (TG800FCAB) and a dry pump (NeoDry30E). The base pressure of $1 \times 10^{-5}$ Pa has been achieved up to now.

The diffractometer mainly consists of $2\theta$, $\theta$, and $\tau_z$ stages as shown in Fig. 2(a), and the specifications are listed in Table 1. The horizontal plane is the scattering plane of this diffractometer. In soft x-ray region, the wavelength is one order longer than that of hard x-ray. Hence one order lower angle resolution is enough for the experiments. In general two-circle diffractometer, the tilt of sample is aligned to set properly a scattering vector with respect to the scattering plane. By changing the height of the detector using the $\tau_z$ stage, on the other hand, this diffractometer can detect the signal out of the scattering plane without an alignment of sample tilt. This experimental configuration gives us new option of the measurements.

The sample position can be aligned by three orthogonal translation, $x_s$, $y_s$, and $z_s$, stages. In temperature dependence experiments, the sample position moves owing to the thermal expansion of cold head of cryostat. In organic materials, radiation damage often becomes a problem. The beam position on the sample should be changed depending on the irradiation time. Moreover, the stroke of $z_s$ stage is large enough to set several samples on the cryostat, and then the sample for measurement can be changed automatically using the $z_s$ stage.
Figure 2. Schematic view of (a) diffractometer and (b) alignment base. The movements of rotation and linear stages are illustrated.

Table 1. Specification lists of diffractometer and alignment base.

| Specifications | $2\theta$ | $\theta$ | $\tau_z$ | $x_s$ | $y_s$ | $z_s$ |
|---------------|-----------|----------|----------|-------|-------|-------|
| Travel range  | $360^\circ$ | $360^\circ$ | 80 mm | 25 mm | 25 mm | 200 mm |
| Resolution    | 0.0003$^\circ$ | 0.0003$^\circ$ | 1 $\mu$m | 0.1 $\mu$m | 0.1 $\mu$m | 0.2 $\mu$m |
| Repeatability | < 0.0005$^\circ$ | < 0.0005$^\circ$ | < 2 $\mu$m | < 1 $\mu$m | < 1 $\mu$m | < 1 $\mu$m |

| Specifications | $X$ | $Z1$, $Z2$, $Z3$, $Z4$ |
|---------------|-----|-----------------------|
| Travel range  | 50 mm | 200 mm |
| Resolution    | 0.4 $\mu$m | 0.1 $\mu$m |
| Repeatability | < 0.5 $\mu$m | < 0.5 $\mu$m |

2.2. X-ray detecting system

In the RXS experiment, the fluorescence from the sample usually becomes main background. Hence the reduction of background is important to detect the weak RXS signal reflecting the change of physical property. First, the cross slit screen with four blades is placed between the sample and the detector. [3] The intensity of the fluorescence is proportional to the solid angle of detecting area of detector, while the Bragg spot has a sharp profile. The slit is used to tune the solid angle in order to improve the signal-to-noise ratio. Next a Silicon drift detector (SDD) with good energy resolution (about 100 eV) is installed. The fluorescence with energy far from the incident X-ray energy can be completely eliminated by using the SDD. By using this system, we succeeded in observing a weak RXS signal in several experiments. Even in x-ray absorption spectroscopy (XAS) measurement, the signal-to-noise ratio is remarkably improved, since the fluorescence from other elements can be excluded by the SDD.
2.3. Cryostat
He-flow type cryostat is installed for the low temperature experiment, and achieves the cold head temperature from 300 K to <10 K. However, radiation shield, which is generally used in low temperature experiments, cannot be utilized in soft X-ray region. Because the incident and scattered X-rays are also absorbed by the radiation shield. Hence the sample surface is heated up by the radiation, and it is difficult to estimate the real sample temperature. By using this cryostat and the diffractometer, the transition temperature less than 20 K could be detected up to now.

2.4. Alignment base
In order to align the diffractometer within the synchrotron beam, the alignment base are used as shown in Figs. 1(a) and 2(b) Generally, the alignment can be performed by two linear (X, Z) and two rotational (Rx and Rz) dimensions. The base has the X and Z stages, and the Rz rotation is virtually performed using Z1, Z2, Z3, and Z4 axes. The Rx rotation is substituted by the 2θ rotation.

3. Future prospects
In RXS experiments, polarization and azimuthal angle dependences become a key information especially to determine the symmetry of ordered orbital and the direction of magnetic moment. Moreover, the charge, orbital, and spin ordered states are coupled each other in SCES. The RXS signal generally contains multiple components reflecting charge, orbital, and spin states and local crystal structures. The RXS components can be separated utilizing the polarization and azimuthal angle dependences. Therefore, we plan to install a sample rotating stage and a polarizer. The analyzer stage can be mounted, taking advantage of the large vacuum chamber.

4. Conclusion
In order to perform the RXS measurement in soft X-ray region, we have developed new in-vacuum diffractometer and the related systems. The diffractometer was installed in the beamlines, BL-16A (300~1500 eV) and BL-11B (1.7~5.0 keV), at the Photon Factory, and was commissioned. Utilizing the developed diffractometer, not only the RXS but also the XAS measurements were carried out to elucidate the electronic states, and the ordering of the charge, spin, and orbital states in SCES. [4, 5, 6]

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[1] Matsumura T, Nakao H, and Murakami Y 2013 J. Phys. Soc. Jpn. 82 021007
[2] Beale T A W, et al. 2012 Eur. Phys. J. Special Topics 208 89
[3] Okamoto J, Horigane K, Nakao H, Amemiya K, Kubota M, Murakami Y and Yamada K 2013 J. Phys.: Conf. Ser. 425 202003
[4] Okamoto J, Horigane K, Nakao H, Amemiya K, Kubota M, Murakami Y and Yamada K 2011 J. Electron Spectrosc. Prlat. Phenom. 184 224
[5] Takahashi R, Okazaki R, Yasui Y, Terasaki I, Sudayama T, Nakao H, Yamasaki Y, Okamoto J, Murakami Y, and Kitajima Y 2012 J. Appl. Phys. 112 073714
[6] Y. Takahashi, H. Nakao, R. Kumai, S. Ishibashi, S. Horuchi, M. Kohyama, K. Kobayashi, Y. Yamasaki, J. Okamoto, T. Sudayama, Y. Murakami and Y. Tokura, to be submitted to J. Phys.: Conf. Ser.