Construction of a soft X-ray diffractometer with a 7.5-T superconducting magnet

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Abstract. We have constructed a soft X-ray diffractometer with a 7.5-T superconducting magnet in order to study ordered electronic structures under high magnetic field. In this paper, we present the features of this system and an example of resonant soft X-ray scattering measurement under the magnetic field of (LaMnO3)3(SrMnO3)3 superlattice with a remarkable negative magnetoresistance effect.

1. Introduction

Resonant soft X-ray scattering (RSXS) is a powerful method for investigating ordered electronic structures in strongly correlated electron systems (SCES). For example, RSXS at transition-metal (TM) L2,3 edges of 3d TM compounds is much more sensitive to the magnetic structures formed by TM 3d electrons than the resonant X-ray scattering (RXS) at the TM K edge, since the TM L2,3-edge RSXS directly measures the TM 3d states through the 2p to 3d transition, whereas the TM K-edge RXS measures the TM 4p states through the 1s to 4p transition [1]. Recently, the external-field effect has become an important issue concerning ordered TM 3d electronic structures, since novel physical properties, such as multiferroics [2] and colossal magnetoresistance [3], are observed at the boundaries of various electric and magnetic phases, and are very sensitive to the external field. Therefore, it is required to measure RSXS under various external conditions besides temperature. Magnetic field is an important parameter since multiferroics and colossal magnetoresistance are closely related with magnetic structure and some of them show metamagnetic transition under high magnetic field [4]. Most previous RSXS measurement systems, however, can apply only a low magnetic field below 1 T by using a permanent magnet and/or electromagnet; no RSXS measuring systems with a superconducting magnet (SCM) have been used to apply a high magnetic field. Since light in the soft X-ray (SX) region is strongly absorbed, these RSXS systems need to be set in a high vacuum (HV) chamber.
2. Instruments

Figure 1. (a) Appearance of the SCM-RSXS system. (b) Schematic view of the diffractometer and the SCM. The magnetic field is applied orthogonal to the scattering plane.

Figure 2. (a) Inside the vacuum chamber. The red broken curve shows the scattering plane. (b) A cross-section of the SCM at the scattering plane. Shaded areas show the regions screened by the thermal shields, supports, and the liq. He vessel. (c) Appearance of the SCR stage.

We have constructed a SX diffractometer with a 7.5-T superconducting magnet in HV (SCM-RSXS), in order to measure the RSXS signal reflecting ordered electronic structures under high magnetic field. Figure 1(a) shows the appearance of the SCM-RSXS system. This system has a large vacuum chamber in order to install the SCM and other devices such as slits and detectors in HV. The base pressure of the chamber is $1 \times 10^{-5}$ Pa. Figure 1(b) shows the schematic view of the diffractometer and the SCM.
Samples are cooled down by the He-flow cryostat and the lowest temperature is 8 K at the sample holder. Scattered light from the sample is detected by a silicon drift detector (SDD) with good energy resolution of about 100 eV. In front of the SDD detector, a four-blade slit is placed in order to measure weak RSXS signals of superstructures by subtracting backgrounds such as fluorescence [5]. This diffractometer has a quasi $\chi$ axis called tau in order to detect the signal out of the scattering plane without changing the sample tilt. The effective range of $\chi$ is $\pm 2.5^\circ$. The scattering plane is set between the two coils and the SCM applies the magnetic field orthogonal to the scattering plane up to 7.5 T. Magnetic field applied upward is defined as positive. The SCM has a large 13-liter liq. He vessel to cool the coils, and the SCM can be used for about 30 hours without adding liq. He, even at the magnetic field of 7.5 T.

Figure 2(a) shows the arrangement of devices inside the vacuum chamber. The red broken curve shows the scattering plane. The SCM has windows to transmit the incident and scattered light toward the sample and the detector, respectively, but there are blind areas in the scattering plane. Figure 2(b) shows the cross-section of the SCM at the scattering plane. Shaded areas show the regions screened by the thermal shields, supports between the two coils, and the liq. He vessel. In order to compensate the 20 regions, we rotate the SCM itself around the same rotation axis of $\theta$ and $2\theta$, called SCR, as shown in Fig. 2(c). Finally we cover the 20 range from 0° to 165° in the SCM-RSXS system; the 20 region above 165° is limited by the slit.

3. Experiment and Results

Superlattices formed by the heterostructures of strongly-correlated insulating oxides have been fascinating materials since the discovery of superconductivity at the interface between SrTiO$_3$ and LaAlO$_3$ [6]. Among them, superlattices composed of antiferromagnetic insulators LaMnO$_3$ and SrMnO$_3$ layers are of particular interest, since a wide variety of electronic and magnetic phases depending on the stacking structures have been reported [7]. These LaMnO$_3$-SrMnO$_3$ superlattices are fabricated on LSAT (001) substrate by pulsed laser deposition, since lattice matching between the layers and the substrate is also crucial to the physical properties of the superlattices [7,8]. The Mott insulator (LaMnO)$_3$(SrMnO)$_3$ superlattice (denoted as L3S3) is located at the boundary of the antiferromagnetic-insulator and ferromagnetic-insulator phases and shows a remarkable negative magnetoresistance effect under the magnetic field of 7 T at 10 K [7]. Namely, ferromagnetic-metal (FM) phase is expected to exist at 7 T. In order to study the Mn 3d electronic structures which form the magnetically ordered structures in the expected FM phase, we measured the RSXS for L3S3 at the Mn $L_{2,3}$ edge by using the SCM-RSXS system.

The RSXS measurement was done at the SX undulator beamline BL-16A in the Photon Factory, KEK. We applied a magnetic field of $\pm 7$ T orthogonal to the scattering plane; the magnetic moment of L3S3 is expected to be nearly orthogonal to the scattering plane. In order to measure the resonant magnetic scattering component effectively [1], we set the electric-field vector of the incident linearly polarized light in the scattering plane (\(\pi\) polarization), as shown in the inset of Fig. 3(b).

For the ferromagnetic structures in L3S3, Mn 3d magnetic moments in LaMnO$_3$ layers are considered to mainly contribute to the ferromagnetism, as shown in Fig. 3(a). In this model, magnetic structures are observed at the 001 reflection. The reflection was indexed using the unit cell of the superlattice whose lattice constant is about 22.8 Å.

Figure 3(b) shows the Mn $L_{2,3}$-edge RSXS spectra of the 001 reflection for L3S3 under the $\pm 7$ T magnetic field at 10 K. Comparing with the XAS spectrum, there are strong resonant structures at the Mn $L_{2,3}$ edges in the RSXS spectra of the 001 reflection. In addition, RSXS spectra of the 001 reflection take different up-and-down line shapes at the Mn $L_{2,3}$ edge under the $\pm 7$ T magnetic field as shown in Fig. 3(b). We consider that this difference originates from the reversal of magnetic moment under the positive and negative magnetic field. The scattering intensity of the 001 reflection contains Thomson scattering, reflecting the superlattice structure of L3S3 in addition to the resonant magnetic scattering. When the magnetic moment of L3S3 is reversed by reversing the external magnetic field, the sign of the resonant magnetic scattering of the 001 reflection also reverses [1], although the sign of
Thomson scattering of the 001 reflection does not change. Then, the difference under the positive and negative magnetic field appears at the 001 reflection via the interference between Thomson scattering and resonant magnetic scattering.

![Figure 3](image)

**Figure 3.** (a) Schematic view of the (LaMnO$_3$)$_3$(SrMnO$_3$)$_3$ superlattice. Red arrows show the magnetic moments in LaMnO$_3$ layers. (b) Mn $L_{2,3}$-edge RSXS spectra of the 001 reflection for (LaMnO$_3$)$_3$(SrMnO$_3$)$_3$ superlattice measured under the ±7 T magnetic field at 10 K with $\pi$ polarization. XAS spectrum of the superlattice measured with FY-mode at room temperature. Inset shows the experimental setting.

4. Conclusions
We have constructed a soft X-ray diffractometer with a 7.5-T superconducting magnet in order to perform resonant soft X-ray measurements under high magnetic field. We measured the Mn $L_{2,3}$-edge RSXS spectra of the 001 reflection for the (LaMnO$_3$)$_3$(SrMnO$_3$)$_3$ superlattice in the ferromagnetic-metal phase under a ±7 T magnetic field, and observed the difference of RSXS spectra corresponding to the reversal of the Mn 3$d$ magnetic moment.

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