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Introduction of photoemission electron microscopes at SPring-8 for nanotechnology support

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Abstract

Two photoemission electron microscopy (PEEM) systems with different characteristics have been introduced in SPring-8 for a nanotechnology support project. One is an easy to use system (ELMITEC PEEMSPECTOR), which is equipped with an electrostatic lens. The other one is a high end system spectroscopic photoemission and low energy electron microscope (ELMITEC LEEM/PEEM III), which is equipped with a magnetic lens and an energy filter. Test experiments have been done using the PEEM systems and high quality x-rays at SPring-8. In this paper, some experimental results will be presented.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Nanoscience and nanotechnology have grown worldwide to be important areas in the field of materials research and development in the past few decades, and this trend is expected to strengthen. It has already been verified by preceding PEEM research at low energy third-generation SR facilities such as ELETTLA [1] and ALS [2] that the combination of PEEM with SR widens the application field for PEEM and provides powerful methods for research and development of nanoscience and nanotechnology. Since the beginning of 2002, SPring-8 has begun to be involved in nanotechnology support in the framework of the Nanotechnology Support Project administered by the Ministry of Education, Culture, Sport, Science, and Technology (MEXT) of Japan. In the case of the introduction of PEEM systems into SPring-8, however, it should be noted that there are several intrinsic disadvantages, because of the high energy rings. That is, (i) photoionization cross sections generally decrease, (ii) transmission of

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the lens system decreases, and (iii) image degradation due to chromatic aberrations increases as photon energy increases. Besides this, the available photon fluxes at the VUV-SX beamlines in the high energy rings are generally weaker than those in the low energy rings. Thus we need a clear strategy for the introduction of PEEM systems to SPring-8 to overcome the above-mentioned disadvantages, making use of the particularities of the high energy ring beamlines.

2. The PEEM research field at SPring-8

SPring-8 is the largest third-generation synchrotron radiation facility in the world, with a high acceleration energy of 8 GeV. The beamlines are mainly dedicated to hard x-ray use; however, there are four active soft x-ray (SX) beamlines. These SX beamlines are unusual among the various VUV-SX beamlines of the third-generation SR facilities in having the following characteristics:

(1) Each of the beamlines is equipped with a Hettric–Underwood-type monochromator [3] with a resolving power higher than 10^4 and typically covers the energy range from 250 to 3000 eV. The best performance is obtained in the range of photon energy below 1 keV. This useful photon energy region covers the K edges of elements C, N, and O, L edges of 3d transition metal elements, and M edges of 4f rare earth elements. The typical photon flux is 10^{10}–10^{11} s^{-1} in this energy range, and focusing of the beam down to 10 μm (vertical) $\times$ 50 μm (horizontal), by means of mirrors, is possible due to the low emittance characteristics of SPring-8.
(2) Every beamline has a different type of undulator, with a polarity switching capability. Among them, BL25SU is equipped with twin helical undulators [4], and the helicity of the SX radiation can be switched at the speed of 1–10 Hz [5]. BL17SU (RIKEN/SPring-8), the newest SX beamline in SPring-8, is equipped with a multimode undulator constructed with a combination of electromagnet and permanent magnet arrays [6]. This brand new undulator is designed to produce SX photons with left and right circularly and also horizontal and vertical linearly polarized light. The designed polarity switching frequency range is from 1 to 10 Hz. The high speed polarity switching enables us to capture PEEM image differences due to circular and/or linear dichroism synchronously with polarization switching.

In order to make use of these characteristics, we identify the dominant PEEM research fields at SPring-8 as follows:

(1) Magnetic domain imaging of materials consisting of 3d transition metal elements and/or 4f rare earth elements by means of magnetic circular dichroism (XMCD) and/or magnetic linear dichroism (XMLD).
(2) Chemical imaging by using the resonance enhancement of photoelectrons at core absorption edges. This is considered to be very useful for surface and interface studies including dynamical studies of surface chemistry.
(3) Nanospectroscopy using XANES (x-ray absorption near edge structure), XAFS (x-ray absorption fine structure), and XPS (x-ray photoemission spectroscopy) for bulk and surface nanostructures.

These spectroscopic methods are expected to provide us with electronic, magnetic, chemical, and structural properties for regions of several hundred nm. Expected research targets include: thin film materials for magnetic memories and spin electronic devices; nanostructures fabricated by self-organization or high precision processing in thin films, solid surfaces, and interfaces. Dynamical studies of surface chemical reactions may be an interesting

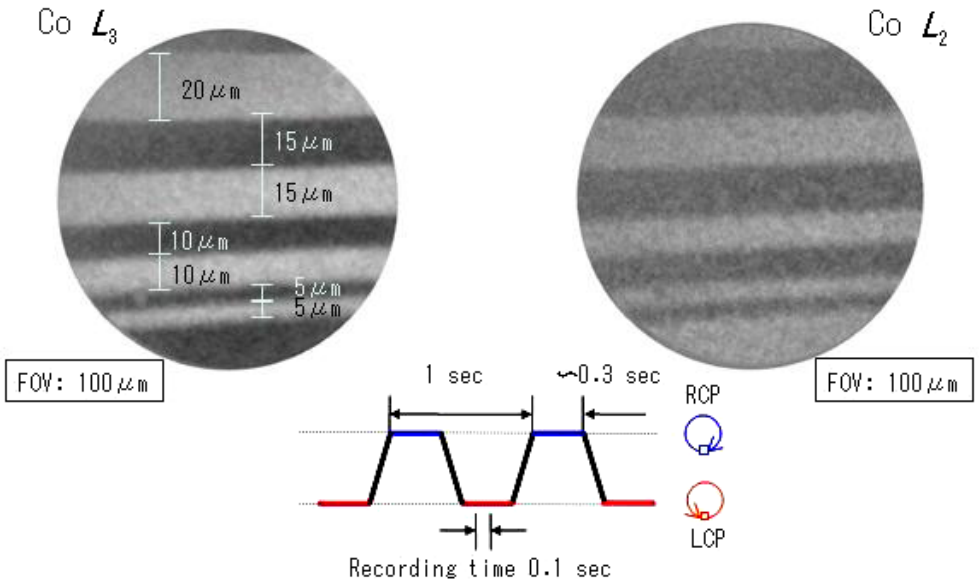

Figure 1. Magnetic domain images of CoCrPt thin film at Co L edges measured by combining PEEMSPECTOR with XMCD. The image contrast of L_3 and L_2 is properly inverted.

research field. Magnetic domain structure change due to pulsating external perturbation utilizing synchronous detection with SR pulses also represents a promising technique for future development.

In order to facilitate effective nanoscience and nanotechnology, it is necessary to realize ease of use for the PEEM experimental apparatus for the benefit of inexperienced users. At the same time, a PEEM system with the highest performance must be constructed in order to make full use of the excellent characteristics of SPring-8 SX beamlines. These two demands cannot possibly be met by a single system, because high performance inevitably requires complicated construction of the system. Thus we decided to introduce two systems; one is designed for versatility and easy use; the other is aimed at achieving the highest performance.

3. PEEM systems and performances

3.1. The easy to use system

We selected PEEMSPECTOR as the easy to use and versatile PEEM experimental station. The PEEMSPECTOR equipment, which consists of an electrostatic lens and a set-up consisting of a deflector and stigmator, is the smallest commercial PEEM in the world, with simple operation. The spatial resolution is guaranteed as 35 nm for mercury short arc lamp excitation. The field of view can be tuned in the range 5–500 μm. The experimental system consists of a sample load lock, a small preparation chamber, and an analysis chamber. The preparation chamber is equipped with reflective high energy electron diffraction (RHEED), an evaporator, an ion bombardier, and an annealing stage. The analysis chamber has a sample goniometer, with XYZ and two-tilt-angle adjustable mechanisms. The goniometer is also capable of rotating the samples by 360° along the surface normal. The sample stage is grounded so that drain current measurement is possible with a standard electrometer. The sample holders are ELMITEC standard. The compact design of the system allows us to move it to several different beamlines to utilize the versatility of the system.

Figure 1 shows a PEEM-XMCD image of perpendicular magnetization recorded domains of CoCrPt thin films [7] at Co L edges studied using PEEMSPECTOR at BL25SU. At BL25SU, five kicker electromagnets were installed in the free spaces of the 4.5 m straight section. The kicker electromagnets can be selectively activated to provide helical light on one optical axis, with oppositely helical light being emitted in a deviated direction to be absorbed by water cooled apertures. Tuning the circularly polarized light to the Co L_3 absorption edge 778.1 eV, two images produced with left (LCP) and right (RCP) circularly polarized light were captured and accumulated synchronously with helicity switching at 1 Hz. At 1 Hz, about 0.1 s was needed to accumulate signals for either circular polarization of the light. Nearly 15 min were taken to obtain the magnetic domain image of CoCrPt. The contrast in figure 1(a) is extracted as $A = (I_{\mathrm{LCP}} - I_{\mathrm{RCP}})/(I_{\mathrm{LCP}} + I_{\mathrm{RCP}})$, where I_{LCP} and I_{RCP} correspond to the images for left and right circular polarization, respectively. The film was prepared to produce a line pattern, and was observed with the linewidths of 20, 15, 10, 5 μm. Setting the photon energy to the Co L_2 absorption edge at 793.2 eV, the same procedure generates a Co L_2 edge XMCD image as shown in figure 1(b). It is recognized that the image contrast of L_3 and L_2 is properly inverted, evidencing that the image corresponds to magnetic domain contrast produced by MCD. This synchronous image capturing method, which is realized via the fast helicity switching capability [5] at the twin helical undulator, is essential for magnetic domain investigation of small MCD materials such as dilute magnetic systems. This is because the helicity switching method can cancel the instrumental drifts occurring during long accumulation periods, necessary to detect weak MCD image differences.

Besides being used at BL25SU, PEEMSPECTOR has also been moved to BL23SU, which is equipped with an APPLE II type of undulator [8–10], to make use of linearly polarized SX photons. Another novel usage of this system has been tried: to realize XMCD recorded domain structure imaging of perpendicular magnetization in CoCrPt thin films at Pt L edges, at the hard x-ray (HX) beamline BL39XU. Measurements at BL39XU verified that applications of PEEM at HX beamlines are quite promising because of the bulk sensitivity. Various experiments have recently been done, including micro-XAFS ones, revealing the versatility of HX excited PEEM observation. These results will be published elsewhere.

3.2. The high end system

We adopted a SPELEEM (spectroscopic photoemission and low energy electron microscope), which is modified from the standard horizontal configuration to an upright configuration, for the high end system (figure 2). The incidence angle from the surface normal of the sample is 74°. The spot size is enlarged by a factor of 3.6 along the vertical direction of the incident beam, which makes the illumination area almost square for the horizontally elongated beam from a SX beamline. The sample can be cooled down to 150 K by a nitrogen flow. Three techniques are used for heating the samples: using radiation, bombardment, and direct heating. The energy analyser allows selection of electrons within a narrow energy window, as low as 0.5 eV, using the energy slit. This reduces the chromatic aberration and makes it possible to measure a nanoscale area x-ray photoelectron spectrum. Element specific imaging is also possible. Rotation around the sample surface normal is sometimes necessary, especially for magnetic domain imaging. We employ a differentially pumped rotary platform (DPRF) with double-differential pumping stages. The DPRF can be rotated by 360° continuously in either direction. The sample is biased at −20 kV with respect to the ground level; thus a picoammeter allowing DC current measurement is employed in a floating configuration for drain current measurement. Drain current measurement is used for obtaining inner core absorption spectra, and is also helpful for aligning the sample with the soft x-ray beam. In the sample preparation

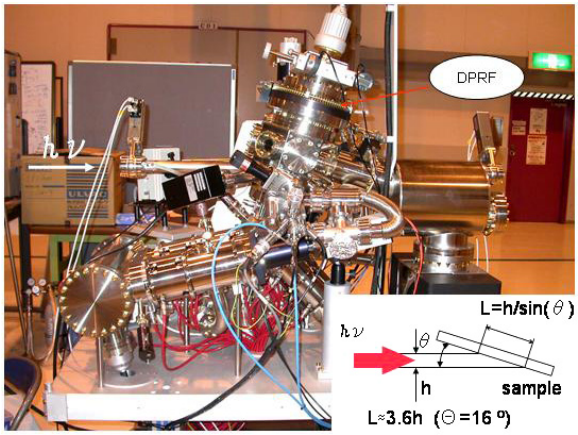

Figure 2. A side view of the SPELEEM introduced in SPring-8. The vertical configuration and DPRF are the key features.

chamber, a sputtering gun is set up for surface cleaning. Two electron beam heated evaporators (EFM) are provided in the main chamber. It is possible to change or reload the evaporation sources without breaking the vacuum of the main chamber, by retracting the evaporator and separating it from the main chamber using gate valves.

We performed the first experiments using this system at BL27SU [11]. This beamline is equipped with a figure-of-eight undulator, which provides SX photons with both vertical (p) and horizontal (s) polarizations [12]. The maximum photon flux is about 10^{11} s^{-1} between 800 and 1200 eV. We investigated the formation and chemical states of Co on Si(111) 7 × 7 surfaces.

Co/Si(111) surface phase structures are interesting for study because of the colourful phases [13] and applications [14]. We deposited Co onto a Si(111) 7 × 7 surface to observe the growth of $CoSi_2$, and studied the chemical states by means of micro-XANES. Co was deposited keeping the substrate at room temperature. After annealing at 600 °C, a Co layer condenses, and $CoSi_2$ begins to grow with strong faceting. The condensation of $CoSi_2$ was observed in LEEM and MEM images, and the faceting was confirmed by the moving of the LEED spots on changing the incident electron energies. Figure 3 shows the micro-XANES absorption spectrum and an XPEEM image. The XPEEM image was taken with soft x-rays at 780 eV, and the FOV is 30 μm. The protrusion part in the centre of the image relates to condensed $CoSi_2$, which is surrounded by a flat region. The micro-XANES absorption spectrum of the protrusion $CoSi_2$ (box symbol) and surrounding Co layer (circle symbol) are shown. The Co L_3 and Co L_2 absorption edges are observed in the spectrum, and three peaks denoted as A, B, and C are clearly distinguished above the Co L_3 edge. The chemical bonding of $CoSi_2$ was well explained by Van den Hoek *et al*: the sp^3 orbitals interact with Co 4s and 3d orbitals [15]. The three splitting peaks are identified as resulting from the non-bonding E_g state, the antibonding Si(sp^3)–Co(3d) state, and the antibonding Si(sp^3)–Co(4s) state, from the lower energy side, respectively. The non-bonding E_g peak of the flat region is weaker than that of $CoSi_2$, suggesting weaker hybridization between Co and Si in the flat region. The flat region is considered to relate to a chemisorbed Co layer.

Because half-order photons are vertically polarized whereas the first-order photons are horizontally polarized in the figure-of-eight undulator [10], it is possible to select vertical and horizontal polarizations by changing the undulator gap without changing the sample

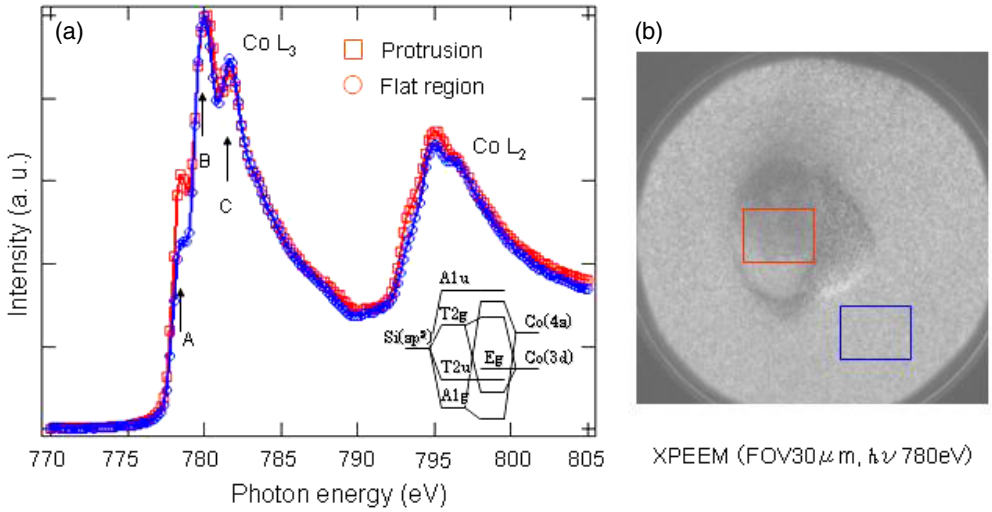

Figure 3. (a) Micro-XANES for $CoSi_2$ and the surrounding flat regions. (b) An XPEEM image of a Co/Si(111) surface.

configuration at BL27SU. Utilizing this capability, we have tried antiferromagnetic domain structure observation for NiO(001) crystal by using XMLD. NiO(001) was cleaved in the open atmosphere, and measurements made without any other cleaning process being carried out. The linearly polarized light is obliquely incident onto the sample, with the glancing angle 16°. The absorption spectrum was first taken by drawing the selected area intensities and photon energies to find the Ni $L_{2,3}$ absorption edges. Two peaks, marked A and B in the upper panel of figure 4, are recognized at the L_2 edge. The peaks were found to show linear dichroism between horizontal and vertical polarizations.

Horizontal and vertical linearly polarized light images with photon energies corresponding to peaks A and B were taken, and the asymmetric images were processed using the formula $A = (I_A - I_B)/(I_A + I_B)$ for both horizontal and vertical polarizations, where I_A and I_B are the images obtained for the two photon energies of peaks A and B, respectively. The bright and dark areas of the images show different antiferromagnetic domains, where the magnetic moment directions are parallel and perpendicular to the polarization vector of the linearly polarized light, respectively. The domain boundaries are found to run along the ⟨100⟩ direction for horizontal linearly polarized light, while they run along the ⟨110⟩ direction for vertical linearly polarized light.

4. Future development

The high end system will be installed into BL17SU, with focusing mirrors, at the end of this year. The designed illumination area on the sample surface is 30 μm × 50 μm, with a photon flux of about 10^{11} s^{-1}. The considerable flux density and polarization switching capability will give us opportunities to perform unique research. As one of the challenges, we are embarking on a new project designed to improve the spatial resolution by using the 'moving focus method' proposed by Ikuta, which has been successfully applied to a transmission electron microscope (TEM) [16]. Adopting this method, we are aiming at sub-10 nm resolution in the high end system at BL17SU with high density SX photon excitation.

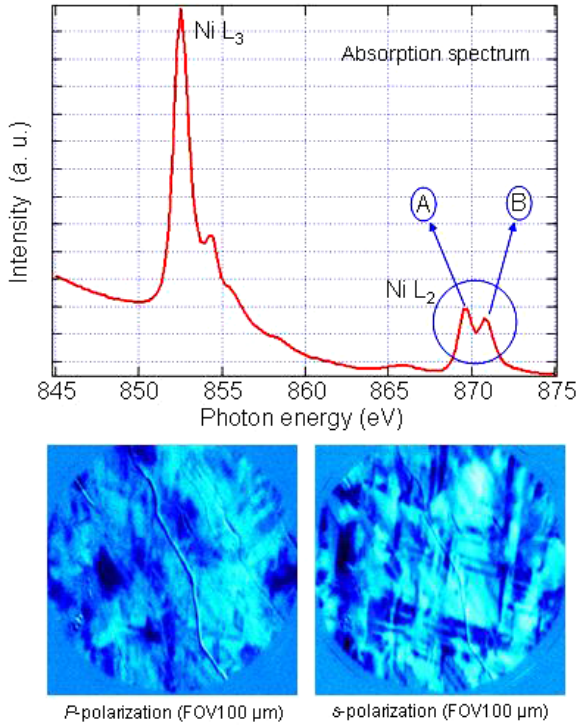

Figure 4. The absorption spectrum of NiO(100) and antiferromagnetic domain images measured by means of the combination of SPELEEM with XMLD. The left and right images show the antiferromagnetic domain images obtained with vertical and horizontal linearly polarized light, respectively.

5. Summary

Two PEEM systems have been successfully introduced in SPring-8 for nanotechnology support. The compact easy to use and versatile system with an electrostatic lens PEEM system is installed at the SX twin-helical-undulator beamline BL25SU, and is going to be used for magnetic imaging and other applications. The high end system is going to be installed into BL17SU, which is equipped with brand new multimode undulators. The challenge of realizing sub-10 nm spatial resolution by cancelling spherical aberration using an image reconstruction method is being tackled.

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