Present status of photoemission electron microscope newly installed in SPring-8 for time-resolved nanospectroscopy

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A photoemission electron microscope (PEEM) system has been newly installed at the soft X-ray undulator beamline (BL17SU) of SPring-8 to realize time-resolved nanospectroscopy for the local transient electronic structures of advanced materials. This PEEM is a versatile machine composed of an electrostatic lens system and is intended for use in specific experiments such as time-resolved measurements. Pump–probe measurements in tandem with a femtosecond pulsed-laser system and an X-ray chopper are now readily available.

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Photoemission electron microscopes (PEEMs) are powerful tools for investigating the electronic states of materials with high spatial resolution.1) In particular, when used in combination with synchrotron radiation (SR) X-rays, PEEM significantly increases the information available, concerning X-ray-absorption spectroscopy (XAS), X-ray-magnetic circular/ linear dichroism (XMCD/XMLD), X-ray photoemission spectroscopy (XPS), and so on.2,3 At SPring-8, a variety of materials have been investigated and a large number of outcomes have been produced.4–10 A spectroscopic photoemission and low-energy electron microscope [SPELEEM (LEEM III, ELMITEC GmbH)] installed at BL17SU (a-branch) is suitable for static image observations with high spatial resolution (~23 nm under the best conditions).11) However, since an electron-extraction voltage of ~20 kV is applied to a sample, the SPELEEM system is not suitable for creating unusual experimental conditions, e.g. applied external fields and time-resolved measurements. This problem can be overcome by complementary use of a versatile PEEM system, wherein an electron-extraction voltage is applied not to the sample stage but to the lens system,12) although its spatial resolution is moderate. In SPring-8, the PEEMSPECTOR (ELMITEC GmbH) machine was utilized as a versatile PEEM. However, its performance deteriorated over 15 years of operation. In this paper, we outline and report on the present status of a newly installed versatile PEEM machine (FOCUS PEEM, FOCUS GmbH).

The FOCUS PEEM [Fig. 1(a)] has been installed at the b-branch (carry-in station Bc) of the BL17SU RIKEN soft X-ray beamline, where circularly and linearly polarized soft X-rays ranging from 0.25 to 1.9 keV (0.48–1.9 keV for vertically polarized X-rays) are available, provided by a multi-polarization-mode undulator.13–15) The soft X-rays with a flux exceeding 1014 photons s−1 are focused down to 10 (vertical) × 25 μm (horizontal) (at an exit slit width of 50 μm) onto the sample. Note that the effective horizontal beam size on the sample plane is doubled because of an incident angle of 30° from the sample plane. The PEEM system with an integral sample stage (IS-PEEM) is a specialty of the FOCUS PEEM, in which a compact sample stage unit is directly attached to the lens unit, thus guaranteeing stability against sample movement relative to the lens. This IS system is convenient for laboratory experiments with fixed research targets; however, to cope with samples of various shapes and experimental conditions such as applied external fields, we independently constructed a high-stiffness six-axis sample manipulator [Fig. 1(b)] instead of adopting the IS system. This manipulator possesses electrically driven axes of translation (X and Y), tilt (tilt-X and tilt-Y), sample-to-lens distance (Z), and in-plane rotation (φ). A robust manipulator body attached to a stiff carved chamber offers a highly stable measurement environment. The spatial resolution of the image using mercury lamp, as estimated from the artificial step edge of a silver film deposited onto a Si substrate was 36.7 nm [Fig. 1(c)]. The achieved resolution was approximately 100 nm when using soft X-rays.

Figures 2(a) and 2(b) show PEEM images obtained in the pre-edge region [1841 eV, Fig. 2(a)] and at the resonant absorption peak [1846 eV, Fig. 2(b)] of the Si K-edge of a gold patchwork pattern fabricated on a Si substrate. Figure 2(c) shows local XAS spectra (photon-energy dependence of image intensities) extracted from two regions of interest (ROIs), “Si” and “Au”, as indicated in Figs. 2(a) and 2(b). The image intensity at the “Si” region clearly increases at the post-absorption edge (a slight increase in the intensity at the region “Au” in the corresponding photon energy is attributed to glare casted from the region “Si” through a phosphor screen or glass flange set just before a CCD camera [note that Au film (~30 nm) is far thicker than the escape depth of photoemission electrons with corresponding photon energy]). Differential-type XPS is also available using an imaging energy filter (IEF, high-pass filter). The lower limit of the kinetic energy of transmissive electrons can be controlled by a bias voltage applied to the sample which is available up to 1600 eV. The upper panel of Fig. 2(d) shows the sample bias dependence of ROI intensities at the region “Au” obtained with a photon energy of 400 eV, the lower panel shows its differentiated profile which is equivalent to a local Au 4f7/2 XPS spectrum. The relation between the sample voltage (Vsample) and the electron-binding energy
\(E_{\text{Bin}}\) is described as \(E_{\text{Bin}} = h\nu - V_{\text{sample}} - \phi\), where \(h\nu\), and \(\phi\) denote the photon energy and the machine potential including the work function, respectively. The full width at half maximum of the XPS spectra was approximately 0.49 eV. Taken into account the photon bandpass \(\Delta E = 5000\) at an exit-slit width of 50 \(\mu\text{m}\) and the natural width of the 4\(f_{7/2}\) core level (\(\sim 0.23\) eV),\(^{16}\) the energy resolution of the apparatus is roughly estimated to be 0.425 eV.

Figures 2(e)–2(l) shows PEEM images of the Ni\(_{81}\)Fe\(_{19}\) circular-dot films (diameter: 5 \(\mu\text{m}\)) obtained at the Fe L\(_{2}\)- and L\(_{3}\)-edges with right- (\(h\nu^+\)) and left-circular (\(h\nu^-\)) polarization X-rays. For beamlines in which quick helicity switching between \(h\nu^+\) and \(h\nu^-\) is unavailable, XMCD-PEEM images obtained by the conventional procedure, \((I_{h\nu^+L3}/I_{h\nu^-L3})\), where first and second subscripts denote the X-ray helicity and the photon energy, respectively, often accompany moiré noises [e.g. Fig. 2(l)] when each constituent image is taken with a large time lag and drift-correction treatment is required. On the other hand, division of a pair of images obtained by quick monochromator switching between the Fe L\(_{1}\)- and L\(_{3}\)-edges [Figs. 2(i) and 2(j)] serves to compensate the periodical inhomogeneity of the microchannel plate. Then, the consequent XMCD-PEEM image, obtained by an image operation of \((I_{h\nu^+L3}/I_{h\nu^-L3})/(I_{h\nu^-L1}/I_{h\nu^+L1})\) [Fig. 2(k)], is free of the moiré pattern and clear magnetic vortices are recognized. We checked that XMLD-PEEM imaging is also available in the same manner as XMCD-PEEM measurements by utilizing horizontally and vertically polarized X-rays (not shown).

A pump–probe PEEM measurement system combined with a Ti: Sapphire pulsed-laser apparatus (\(\lambda = 840\) nm, \(\sim 130\) fs width, 1 kHz) and a high-vacuum X-ray chopper\(^{17,18}\) is readily available. The temporal resolution is determined by the pulse width of the SR (better than 60 ps). Figures 3(a) and 3(b) respectively show a timing diagram of the pump–probe measurements and time-dependent PEEM images of the same sample as in Figs. 3(e)–3(l) under excitation by the laser pulse (fluence: 8 mJ cm\(^{-2}\)) obtained at the Fe L\(_{3}\)-edge. An operation mode H [11/29-filling + 1 bunch (5 mA)] is used for this experiment. The moment at which the laser pulse hits the sample \((\Delta t = 0)\) can be determined easily, since slight distortion of the electron image due to space charging is recognized at this time. Even though the space-charging effect is not favorable in the actual analysis, it is confirmed that the resultant images are tolerable for the magnetic domain analysis if the fluence of the applied pulse is at least lower than 20 mJ cm\(^{-2}\) (note that typical fluence required for magnetization control is around 15 mJ cm\(^{-2}\) or lower).\(^{9,19,20}\) Time-resolved PEEM measurements under other types of stimulation sources such as electric pulses or radiofrequencies are also possible by proper selection of cut-out frequencies of the chopper (1, 3.8, 40 kHz or out of use) and operation modes of the storage
ring (for the operation modes in 2018, see Ref. 21). The sample manipulator is equipped with SMA connectors for future development of the dynamic PEEM measurement system under GHz-microwave excitations.

The shape of the manipulator tip is compatible with ELMITEC’s standard sample cartridges, which are capable of heating samples up to $\sim 1300 \text{ K}$ ($\sim 2000 \text{ K}$ for short flashing). A helium/nitrogen-flow cryostat dedicated to low-temperature experiments is under test experiments. Further testing of the $k$-space lens mode combined with the IEF will enable us to observe angular distributions of photoelectrons or the band structures of single-crystal materials. In addition, hard X-ray PEEM experiments are planned by carrying the measurement system in the experimental hutch of a hard X-ray beamline.

In conclusion, a versatile PEEM machine was introduced at BL17SU at SPring-8. This machine is mainly utilized for special experiments, such as measurements under applied external fields or those that are time resolved. In combination with the soft X-rays provided by the multipolarization-mode undulator of BL17SU, XAS-, XMCD-, and XMLD-PEEM measurements as well as pump–probe measurements under pulsed-laser excitations are now routinely usable. Further developments such as a low-temperature experimental system and the construction of user-friendly software, are now in progress.

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