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Abstract

The principle and design of a new photoemission electron microscope (PEEM), which is called Stereo-PEEM, is described here. Stereo-PEEM can display not only the image of microscopic materials but also the angular distribution of high-energy photoelectrons up to about ±60°, which is about 100-fold the acceptance angle of usual PEEM. This wide angle acceptance for high-energy photoelectrons enables the three-dimensional (3D) display of atomic structure as well as the 3D electronic structure of individual micromaterials. The 3D atomic structure of a sample can be observed directly by taking stereophotographs using photons with angular momentum.

Keywords: PEEM; Stereo-PEEM; Display-type spherical mirror analyzer; Stereophotography

1. Introduction

The recent development of the photoemission electron microscope (PEEM [1,2]) has enabled many types of analysis of small regions of surfaces because recent PEEM has an energy analyzer and can carry out photoelectron spectroscopy on individual materials as small as about 50 nm. The chemical composition and electronic states of such materials can be studied using PEEM. The objective lens of usual PEEM accelerates photoelectrons to the entrance aperture of PEEM by applying an electric field of about 20 kV. Low energy photoelectrons (below about 30 eV) emitted at full solid angles can be collected by this acceleration, but only a few degrees can be collected at approximately 1000 eV. Hence, energy-band dispersion analysis is possible only at low kinetic energies. Atomic structure analysis using photoelectrons is carried out by photoelectron diffraction, photoelectron holography or stereophotography [3–5], which requires a photoelectron angular distribution pattern in a wide angle (about ±60°) at a kinetic energy of approximately 1000 eV. Hence, usual PEEM cannot obtain atomic structure information using photoelectrons. The combination of PEEM with a low energy electron microscope (LEEM [6]) enables structural analysis of individual small materials. However, the structure obtained using LEEM is a whole structure consists of many atomic species. The photoelectron method has an advantage that it enables the analysis of structures around specific atomic species by selecting photoelectrons from the specific core level. Moreover, stereophotography has an advantage in that it does not require calculation and can visualize three-dimensional atomic arrangements directly.

Stereophotography [3–5] is a newly developed method of taking stereophotographs of atomic arrangements by measuring the two-dimensional photoelectron angular distribution (PEAD) from a sample excited by circularly polarized soft X-rays. The forward-focusing peaks in PEAD rotate by the same degree as the parallax in a stereo-view. The patterns in wide angle PEAD excited by left and right helicity circularly polarized light are stereopictures as they are. By viewing each left and right picture with the left and right eyes, respectively, one can directly image the...
Fig. 1. Schematic drawing of Stereo-PEEM.

Fig. 2. (a) Schematic drawing of the large acceptance angle objective lens for ±50° collection. EL1, EL2, EL3, and EL4 are cylindrically symmetric lenses. The blue dotted lines are constant potential lines and the red lines are the trajectories of electrons. (b) Parameter of the ellipsoid. (c) Aberration vs ratio $\gamma = a/b$ of ellipsoid. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)
atomic arrangement of the sample. These PEAD patterns can also be used as photoelectron diffraction or holography patterns, which can be used to analyze atomic structure in detail and with high accuracy. A display-type spherical mirror analyzer (DIANA) [7,8] can show these patterns directly on a screen without any computer processing. When we use ultraviolet light as an excitation source we can obtain three-dimensional (3D) band structure and the Fermi surface mapping of valence band as well [9–11]. The 3D atomic arrangement and electronic structure obtained in this way show an average of the structures in the area illuminated by the excitation light beam, which is about 0.3 mm in diameter for the beamline BL25 of SPring-8. It is necessary to put a special lens system in front of DIANA to select a small region in which the 3D atomic arrangement and electronic structure are studied. This is the concept of Stereo-PEEM.

Fig. 1 shows a schematic drawing of this concept of Stereo-PEEM. When the sample is placed at the entrance of DIANA, which is shown by an aperture close to IL4 in this figure, the 3D band mapping and 3D atomic arrangement of the sample are displayed directly on the screen. A spherical electric field between the main grid and the outer sphere focuses photoelectrons with a selected kinetic energy to the exit aperture. These photoelectrons are detected by a pair of microchannel plates (MCPs) and a fluorescent screen. The emission and detection angles are the same for all trajectories, and a nondistorted PEAD of ±60° is directly displayed on the screen. Thus far, a 3D band mapping [9], photoelectron or Auger diffraction and a stereophotograph of atomic arrangement [5], which are shown in the inset of Fig. 1, have been measured. The energy resolution (ΔE/E) of this analyzer is 0.3% and the angular resolution is 0.5° [8].

In Fig. 1 a large-acceptance-angle objective lens and a lens system, which consists of six lenses (IL1, IL2, PL1, PL2, IL3, IL4 here) and two apertures (CA and FA), are placed between the sample and DIANA. Because this figure shows only the concept of the design, the number of the lens and their function and the positions of the apertures are not those in an actual design, which will be shown in Fig. 4. This lens system enables the display of not only PEAD but also images of samples on the screen. Using the field aperture denoted by FA, a small area on the sample, which is shown by a small circle in the inset drawing denoted by “Image”, can be selected. The 3D atomic arrangement and electronic structure of the selected area can be displayed on the screen by changing the operation mode of the lens after FA.

The most difficult and important part of this system is the development of the large acceptance angle objective lens. This objective lens should focus all the electrons emitted from the sample at a solid angle of about ±60°. A typical electrostatic lens has a high degree of spherical

Fig. 3. Off-axis aberration for acceptance angles up to (a) 40°, (b) 30°, (c) 20°, and (d) 10°. The summary is shown in (e). “σ/M” is a quantity of the aberration disk size divided by the magnification ratio, which shows lateral resolution on the sample. “d” is the distance from the axis.
aberration and can focus emitted electrons within only a few degrees, and there has been no lens that can focus electrons emitted at such large angles.

2. Large acceptance angle objective lens

We have successfully developed a small-aberration large-acceptance-angle objective lens [12,13]. Fig. 2a shows a schematic drawing of the large-acceptance-angle objective lens for the ±50° collection. It consists of an ellipsoidal mesh, and four cylindrically symmetric electrostatic lenses, namely EL1, EL2, EL3, and EL4. The potential of the ellipsoidal mesh, EL1 and EL4 are of ground level. The constant potential lines are shown in the figure as dotted blue lines. The trajectories of electrons are shown by thin red lines. All trajectories within ±50° converge to one point with very small aberration. Fig. 2b shows a parameter of the ellipsoid, and Fig. 2c shows a graph of aberration vs the ratio of the long radius a to the short radius b of the ellipsoid. The aberration decreases dramatically when we set the ratio (\( \gamma = a/b \)) to approximately 1.73. The modification of the shape from an exact ellipsoidal shape is important for achieving nearly zero spherical aberration. Although the spherical aberration can be made as small as we want by increasing the number of parameters in modification of the shape and voltages of the mesh and the lenses, the mesh hole’s effect hinders the actual aberration from decreasing to less than 1 μm. The small hole of the mesh produces a modulation of the potential around the hole, which make the focusing spot size equal to or several times smaller than the hole size. A few 10 μm hole’s of the mesh makes the actual aberration to be around or less than 10 μm. It is necessary to use nano-mesh to obtain nanometer resolution. Another factor to increase the aberration is the fabrication error of the mesh shape. An error of 5 μm in fabrication of the mesh shape will produce several μm aberration. Hence today’s technological limitation limits the spherical aberration to be several μm. In the following calculation the mesh hole’s effect is neglected and only the pass energy electrons are considered. Chromatic aberration will be discussed in Section 4.

Fig. 3 shows the off-axis aberration up to ±20 μm for acceptance angles of (a) 40°, (b) 30°, (c) 20°, and (d) 10° limited by the contrast aperture. The degree of aberration increases when the source point deviates from the central axis as shown in Fig. 3a–d. For the acceptance angle of ±40°, the spot size increases to about 2 μm when the distance from the lens axis is 20 μm as shown in Fig. 3a. When the acceptance angle decreases to ±10°, the calculated spot size decreases to much smaller than 1 μm within 50 μm from the lens axis. Note that when the source is on the axis, the blur is much smaller than 1 μm even when the acceptance angle is ±50°. Hence, we estimate that the lateral resolution is less than 0.1 μm (when the mesh hole size is small) for the lens axis even when the acceptance angle is ±50°.

3. Design of lens system for Stereo-PEEM

The total lens system of Stereo-PEEM was designed using a computer simulation of the trajectories and is shown in Fig. 4. Fig. 4a shows a simulation of the imaging mode, which is used to display an image of the sample on the screen, and Fig. 4b shows a simulation of the diffraction mode, which is used to display the angular distribution of photoelectrons on the screen. In both cases, the lens configuration is the same and only the voltages after the field aperture are different. DIANA is used as an energy filter, and is discussed in the next section.

When we observe the image of the sample, we put a contrast aperture at the third diffraction plane as shown in Fig. 4a. A small contrast aperture can be used to limit the acceptance angle within about ±10° to obtain a wide and clear image.

When we observe the PEAD pattern from a specific area of the sample, that area should be moved to the lens axis. Then, a small field aperture is inserted to select the area. Next the contrast aperture is removed to obtain the entire angular distribution from that area.

An example of a simulation of PEAD is shown in Fig. 5. Fig. 5a shows an image of 200-μm-size area taken with a contrast aperture (CA) of ±10° acceptance angle. A field aperture is placed at the center as shown in Fig. 5a. Fig. 5b
shows the PEAD pattern when a 300-μm FA is used at a 30-times-magnification plane as shown in Fig. 4b. This figure shows that the PEAD from a 10-μm-size area is clearly seen. When a 30-μm field aperture is used, the PEAD from a 1-μm-size area can be seen more clearly.

At present, we have succeeded in obtaining the first image from a large acceptance angle objective lens and found that this objective lens works according to the simulated results.

4. Energy resolution and chromatic aberration

DIANA is used at a symmetric configuration in Fig. 1 but it is used at an inclined configuration in Fig. 4. The principle of energy filter in the symmetric configuration is high-pass and low-pass filter using many obstacle rings and retardation grids [7]. The energy resolution in the inclined configuration is sufficiently high even if we do not use obstacle rings and retardation grids [14,15] because the energy filtering is made utilizing a dispersion property. The inclined configuration can be adopted when the angular divergence is small. The angular divergence in Fig. 4b is ±8°.

The energy resolution is about 1% of the pass energy in DIANA when the aperture size is 2 mm. The lens just before the entrance aperture is a deceleration lens and reduces the kinetic energy to one tenth of the initial kinetic energy. Hence 2 mm aperture corresponds to a total energy resolution of $10^{-3}$ and 0.2 mm aperture corresponds to $10^{-4}$.

In the imaging mode of Fig. 4a the spot size at the entrance aperture is 2 mm. In this case the entrance aperture is a diffraction plane, and the full spot size corresponds to the initial angular divergence of ±50°. Hence the total energy resolution is $10^{-3}$ for the initial angular divergence of ±50° and $10^{-4}$ when the angular divergence is limited to ±5°. This limitation is done by the contrast aperture or the entrance- or the exit-aperture.

In the diffraction mode of Fig. 4b the spot size at the entrance aperture is 300 μm when the field aperture of 300 μm is used. In this case the entrance aperture is an image plane, and the full spot size corresponds to the detecting area on the sample of 10 μm. Hence the total energy resolution is $10^{-4}$ for the detecting area on the sample of 7 μm and $10^{-5}$ when the detecting area is limited to less than 1 μm. This limitation is done by the field aperture or the entrance- or the exit-aperture.

The chromatic aberration $\delta_c$ of this system is in proportion to the acceptance angle and the total energy resolution. When the acceptance angle is ±50° the chromatic aberration $\delta_c$ is about 5 and 0.5 μm for the total energy resolution of $10^{-4}$ and $10^{-5}$, respectively. The overall aberration of this system is much larger than that of usual cathode objective lens system because of the large acceptance angle at high energies.

5. Conclusion

We showed that the combination of a large acceptance angle objective lens and a lens system can be used to realize Stereo-PEEM with large acceptance angles even at high kinetic energies. The PEAD from a selected microscopic area can be obtained using a field aperture. When we use circularly polarized light for excitation, PEADs obtained are stereophotographs of atomic arrangement of the sample. In this way, Stereo-PEEM can obtain three-dimensional atomic and electronic structures of microscopic-materials. Although the spatial resolution is around μm range, the unique performance to obtain wide-angle angular distribution at high kinetic energies is useful for structure analysis around a specific atom using photoelectron diffraction or stereograph.

Recently soft X-ray synchrotron beams can be focussed to sub-micron dimensions. With such a system one can obtain higher spatial resolution without the need for the complex electron optical system described here. The system described here has an advantage for μm range object that the analysis can be made using usual laboratory X-ray source such as Al Kα rather than special synchrotron
beams. The image of the sample can be taken without moving the sample or the light beam. Hence this system can be used in laboratory conveniently.

References