Principle and basic design of omnidirectional photoelectron acceptance lens

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We propose a ±90°-acceptance spherical aberration-corrected electrostatic lens based on the cathode lens technique used in photoemission electron microscopy. This lens, which we call “omnidirectional photoelectron acceptance lens (OPAL)”, is aimed at realizing 2π-steradian photoelectron spectroscopy in a wide energy range. For this lens, modifications of a simple cathode lens were studied in detail by ray-tracing calculations. Then, modified cathode lenses were combined with a decelerating mesh lens in order to achieve a focusing lens with a full acceptance angle of ±90°. Some basic designs of the lens are presented. These designs allow for 2π-steradian photoelectron spectroscopy not only in the UPS regime, but also in the XPS regime, which may open new horizons in photoelectron spectroscopy.

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1. Introduction

Atomic-level investigations of materials concerning unusual properties such as high-temperature superconductivity, topological surface states, colossal magnetoresistance, etc. will give keys to discovery leading to innovation in technology. In such investigations, it is important to determine both atomic and electronic structures at surface, bulk or interface when focusing only on limited information. However, this should be avoided, because one can easily miss the hidden essential point and make a wrong conclusion focusing only on limited information.

There are various photoelectron spectroscopy-based analysis methods that allow us to access detailed atomic and electronic structures: angle-resolved photoelectron spectroscopy (ARPES),[1–3] momentum microscopy,[4–6] photoelectron holography,[7–10] atomic stereography,[11–13] and diffraction spectroscopy.[14–17] ARPES is a standard method to investigate surface electronic structures. Momentum microscopy is a promising method for band structure imaging and two-dimensional (2D) spin detection from micro- and nanoscale region. Photoelectron holography is a method to determine three-dimensional (3D) atomic arrangement around a specific atom. Atomic stereography, which uses X-ray circular dichroism, is a method for direct recognition of 3D atomic arrangement. Diffraction spectroscopy is an atomic-site selective photoelectron spectroscopy method using photoelectron diffraction, which allows us to access local atomic and electronic structures.

In conventional ARPES, a concentric hemispherical analyzer (CHA) is used to measure energy and angular dependence of photoelectron intensity. Here an input lens system is used to transport emitted electrons from the sample surface to the hemisphere entrance, where a slit perpendicular to the energy dispersion direction is inserted. In the angular mode, 2D angular distribution is projected at the hemisphere entrance, then the distribution is trimmed by the slit, and 1D angular distribution with energy dispersion is obtained at the detector screen of CHA. Here, 2D angular distribution with high energy resolution can be obtained by step-by-step measurement with sample and/or analyzer rotation. However, given the analyzer with an ordinary input lens, a considerable amount of time is required for measuring wide-range 2D angular distribution, because its acceptance angle is severely limited by the spherical aberration of the lens. Moreover, it should be mentioned that the atomic-orbital analysis using linearly polarized light,[8,19] which is a powerful method to directly determine the spatial orientations of atomic orbitals, is, in principle, not possible by the sample-rotation approach.

To efficiently obtain wide-range 2D angular distribution, there are several different approaches: (i) the use of display-type spherical mirror analyzer (DIANA), (ii) the use of a retarding field analyzer (RFA), (iii) the use of mesh lens, and (iv) the use of the PEEM (photoemission electron microscopy) technique. DIANA[20–22] has a wide acceptance angle of ±50° to ±60° and has been successfully used in the fundamental study of band structure and atomic arrangement. However, it is difficult to achieve high energy resolution in this analyzer; the obtained energy resolution is around 0.5%. A wide acceptance angle of around ±60° is also realized by RFA. The main part of the analyzer is a high-pass energy filter consisting of three or four spherical grids. In this analyzer, it is difficult to measure weak signals buried in large background noise, even with use of a lock-in amplifier. Recently, improved RFA with an energy resolution of around 0.1% or better was developed.[23]

The approach (iii) allows a wide acceptance angle for conventional electron spectrometers such as CHA by correcting the spherical aberration of an ordinary electron lens using a spherical or ellipsoidal mesh electrode.[24–35] The acceptance angle available in this approach is around ±30° for the spherical mesh case[24] and around ±50° for the ellipsoidal mesh case.[25–35] In this approach, both a wide acceptance angle and high energy resolution can be realized in a wide energy range. Here, in contrast with the conventional ARPES approach, it is possible to measure wide-range
2D angular distribution without sample-rotation, but with use of deflector scanning,[34] in which 2D angular distribution formed at the entrance plane of CHA is scanned by a deflector. However, for 2π-steradian photoelectron spectroscopy, step-by-step measurement with sample tilting is still required.

The approach (iv) achieves a full acceptance angle of ±90° using a cathode lens (see Refs. 36, 37 for a detailed discussion on a cathode lens). Here a high acceleration voltage of typically 20 kV or up to around 50 kV is applied between the sample and the entrance of the objective lens. This technique has been used in PEEM[38-40] and related instruments including an energy-filtered PEEM,[41] SPELEEM,[42] NanoESCA,[43] and Momentum Microscope.[44-46] The accelerated electrons are decelerated and energy-filtered by a retarding-grid analyzer[41] or by a band-pass analyzer such as CHA.[44-46,42,43] Here the full acceptance angle is realized for electron kinetic energies $E_k$ up to some tens of eV. However, the acceptance angle in this approach considerably decreases with increasing the electron kinetic energy.

In this paper we propose a combination approach to achieve a full acceptance angle of ±90° in a much wider energy range. Here we combine the above two approaches (iii) and (iv) (the mesh lens and the cathode lens approaches) in a certain manner. Using ray-tracing calculations, we study in detail the spherical aberration nature of a cathode lens. Here we consider deformations of a simple cathode lens to reduce or correct the spherical aberration. Combining a deformed cathode lens with a decelerating mesh lens, we obtain a full acceptance angle spherical aberration-corrected electrostatic lens, which we call "omnidirectional photoelectron acceptance lens (OPAL)". This paper is the first report on OPAL and we describe the basic principles of the approach starting with a description of the conventional PEEM approach.

2. Limitation of the conventional PEEM approach

Figure 1(a) shows a cathode lens consisting of a flat conducting plate (sample) and a plane conducting grid (mesh) set parallel to each other. Electrons are emitted from the origin of the coordinates (the center of the sample) and a high acceleration voltage $V_a$ is applied between the sample and the grid. Then, assuming a uniform acceleration field between the sample and the grid), the trajectories of the electrons emitted with kinetic energy $E_0$ and emission angle $\theta$ are expressed by

$$x = (-v_0^2 \cos \theta \sin \theta + v_0 \sin \theta \sqrt{v_0^2 \cos^2 \theta + 2az})/a, \quad (1)$$

where $v_0 = \sqrt{2E_0/m}$ is the velocity of the electrons at the sample surface and $a = eV_a/2m$ is the acceleration by the field; $m$ is the electron mass and $L$ is the distance between the sample and the grid. The final angle $\theta'$ of the trajectory relative to the optical axis $z$ is given by

$$\tan \theta' = v_0 \sin \theta / \sqrt{v_0^2 \cos^2 \theta + 2aL}. \quad (2)$$

Electrons emitted from the sample are bent by the acceleration field and draw parabolic trajectories, as shown in Eq. (1). This simple plane mesh lens is used to extend the acceptance cone angle of photoelectron spectrometer.[44,45] Notice that a trajectory, expressed by Eq. (1), does not start from the vertex of the corresponding full parabola, except in the grazing-emission case ($\theta = \pm 90°$). The trajectories shown in Fig. 1 [calculated by Eq. (1)] are those with initial angles $\theta$ from −90° to 90° with a step of 10°. Here the initial kinetic energy $E_0$ is set to 1 keV and a voltage of 20 kV is applied to the grid, with the sample being grounded. For the present, we set the distance $L$ to 10 mm.

Tangential lines (virtual rays) are drawn from the trajectory points at the grid, supposing that they are incident to the objective lens following the cathode lens. The axially-crossing points ($z, x$) = ($z_0, 0$) of virtual rays are calculated from Eqs. (1) and (2) as follows:

$$z_0 = -\frac{x}{\cos \theta'}. \quad (3)$$

Fig. 1. (a) Electron trajectories, (b) spherical aberration SA and (c) final angle $\theta'$ calculated in a simple cathode lens. A voltage of 20 kV is applied between the cathode (sample) and the counter electrode (a plane grid). GIP is the Gaussian image plane for the virtual image, where the SA is calculated as a function of the initial angle $\theta$.
It is seen here that the virtual ray for the emission at \( \theta = 90^\circ \) always crosses the optical axis at \( z = -L \) and all virtual rays converge at this point in the limit \( E_0 \to 0 \) or \( a \to \infty \). The Gaussian image plane GIP is defined by \( z = z_c(\theta = 0) \). This plane is always on the right side of the point \((-L, 0)\) and it follows immediately from the differentiation of Eq. (3) with respect to \( \theta \) that the point \((z_c, 0)\) gradually approaches the point \((-L, 0)\) with increasing \( \theta \) from \( 0^\circ \) to \( 90^\circ \). This means that the spherical aberration produced by the cathode lens is always positive. Although virtual rays converge only in the limit \( E_0 \to 0 \) or \( a \to \infty \) in the mathematical sense, they can converge point-like in the case where \( E_0 \) is much smaller than 1 keV for \( V_a = 20 \) kV. Otherwise, large spherical aberration occurs, as shown in Fig. 1(b). Figure 1(c) shows the final angles \( \theta' \) as functions of \( \theta \) for the \( E_0 \) considered in Fig. 1(b). In the case \( E_0 = 20 \) eV, not only the spherical aberration but also the final angle is considerably small and a full acceptance focusing lens is possible for a combination of the cathode lens with a deceleration lens.

A deceleration lens is necessary for energy analysis, since the energy resolution of an electron analyzer becomes worse with increase of the pass energy. However, a deceleration lens typically has a large spherical aberration. In Fig. 2 we consider the combination of the cathode lens with a simple deceleration lens. The acceleration voltage of the cathode lens is 20 kV, the same as in Fig. 1. The same voltage is applied to the left electrode of the deceleration lens and the right electrode is set to zero voltage. Initial kinetic energies \( E_0 \) of (a) 100 eV and (b) 1 keV are considered.
accelerated by the cathode lens are decelerated to the initial kinetic energy $E_0$. It seems that the use of this combination lens in a photoelectron spectrometer allows the full ($\pm 90^\circ$) acceptance angle only when the electron kinetic energy $E_0$ is less than some tens of eV. Here the acceptance angle is defined as the largest angle of incidence with which electrons pass through a slit or aperture of an analyzer. For $E_0 \sim 100$ eV and $E_0 \sim 1$ keV, the acceptance angles of the combination lenses are limited to around $\pm 30^\circ$ and around $\pm 10^\circ$, respectively. We aim at realizing a full acceptance angle electrostatic lens in a wide energy range, extending the energy range up to a few keV.

3. Deformations of a simple cathode lens

We consider deformations of the simple cathode lens shown in the previous section. Figure 3(a) is a cathode lens consisting of a flat conducting plate (sample) and an open-convex counter electrode. Here a plane grid forms the top of the convex shape. Figure 3(b) is a cathode lens that does not use a grid in the counter electrode. In both figures, $L$ and $r$ denote the distance between the sample and the counter plate and the inner radius of the cylinder part, respectively. For the present, they are both set to 10 mm. Then the electrostatic field is determined by the height $h$ of the cylinder part. In the following, the initial kinetic energy $E_0$ and the acceleration voltage $V_{ac}$ are set to $1$ keV and 20 kV, respectively. Figure 4(a) shows electron trajectories calculated in cathode lenses in Fig. 3. The electrostatic field and the electron trajectories are effectively changed by the parameter $h/r$ in the range of 0 to $\sim 1$. Here the spherical aberration decreases with increasing $h/r$, as shown in Fig. 4(b). The spherical aberration in the gridless cathode lens is smaller than that in the simple plane grid cathode lens ($h/r = 0$), and that in the cathode lens with a convex counter electrode [Fig. 3(a)] with $h/r$ less than around 0.4, but the final angles $\theta'$ obtained in the gridless cathode lens are large compared to those obtained in the other lenses, as shown in the lower panel of Fig. 4(b). To obtain a spherical aberration-corrected focusing lens in the combination of a cathode lens and an additional electrostatic lens, both the spherical aberration and the divergence angle in the cathode lens should be suppressed to be small.

Figure 5(a) shows another deformation of the cathode lens shown in Fig. 1(a). Here a convex shape is formed on the cathode. The top of the convex shape is the sample. Figures 5(b) and 5(c) are the combinations of a convex cathode with counter electrodes of shapes in Figs. 3(a) and 3(b), respectively. The radius and the height of the cylindrical part of the convex cathode are denoted by $r_0$ and $p$, respectively. $L$ is the distance between the sample plane and the base plane of the counter electrode. Figures 6(a) and 6(c) show electron trajectories in cathode lenses for some variations of the parameters $r_0$ and $p$ in Fig. 5(a). The spherical aberration for the virtual image decreases with increasing the height $p$ and also decreases with decreasing the
radius $r_0$, as shown in Figs. 6(b) and 6(d), respectively. The final angle $\theta'$ increases with increasing $p$ and also increases with decreasing $r_0$, as shown in the lower panels of Figs. 6(b) and 6(d), respectively. Deformation of the cathode lens in the shape of Fig. 3(a) to the shape of Fig. 5(b) allows considerable decrease in spherical aberration, as shown in Fig. 7(b). Here the solid curve corresponds to the left panel of Fig. 7(a). Adjusting the parameters $r_0$ and $p$, spherical aberration can be reduced to around zero. Moreover, negative spherical aberration can be produced by decreasing the radius $r_0$, while the simple cathode lens considered in Sect. 2 always has positive spherical aberration. Deformation of the cathode lens in the shape of Fig. 3(b) to the shape of Fig. 5(c) is also significant to decrease spherical aberration [see Figs. 4(b) and 7(b)].

4. Omnidirectional photoelectron acceptance lens

We design an OPAL combining a cathode lens in Figs. 3 or 5 with a decelerating mesh lens. The lens is optimized so that its spherical aberration becomes as small as possible. Here the adjusting parameters include shape and position parameters and applied voltages. Figure 8 shows an OPAL using a cathode lens in Fig. 3(a). Here the sample is grounded and 20 kV is applied to the counter electrode for $E_0 = 1$ keV. The decelerating mesh lens is composed of five electrodes EL1, ..., EL5, and an ellipsoidal mesh is given at the first electrode EL1. The same voltage as at the plane grid is applied to the electrode EL1 and the region between the grid and the ellipsoidal mesh is kept field-free. Certain voltages lower than the accelerating voltage are applied to the remaining electrodes, with the final electrode EL5 being set to zero voltage. The sample is irradiated by an X-ray or UV light beam passing through the hole shown in Fig. 8. The angle...
between the sample and the irradiation beam is set to 30°.
Electrons emitted from the sample are accelerated by the cathode lens and then decelerated to the same kinetic energy as at the sample surface. In the OPAL, spherical aberration is reasonably small over the full (±90°) acceptance angle. The divergence angle of electron trajectories at the exit plane of the OPAL is around ±11°. Here the distance between the sample and the exit plane is 400 mm, resulting from the setting of \( L = 10 \) mm. Although various conditions including the mesh shape, the arrangement of electrodes, and the voltages applied to the electrodes were adjusted, the spherical aberration could never be reduced to a small degree simultaneously over the full (±90°) acceptance angle.

Figure 10(a) shows a combination of a cathode lens in Fig. 5(b) with a decelerating mesh lens. In this lens, spherical aberration is finely corrected over the full (±90°) acceptance angle. Figure 10(b) shows a combination of a grid-less cathode lens in Fig. 5(c) with a decelerating mesh lens. The spherical aberration in this lens is much smaller than that in Fig. 9(b), showing the effectiveness of the use of a convex cathode. The divergence angles at the exit plane in Figs. 10(a) and 10(b) are both around ±11°. The distance between the sample and the exit plane in Fig. 10(a) is 388 mm and that in Fig. 10(b) is 408 mm, both resulting from the setting of \( L = 10 \) mm.

The effect of the electron-source size on the performance of OPAL is considered as shown in Fig. 11. The upper panel of Fig. 11(a) shows off-axis aberration for the OPAL shown in Fig. 8. Here electron trajectories starting with initial angles \( \theta = 0° \) to ±90° from five point sources with off-axis distances \( x = -0.05, -0.025, 0, 0.025, 0.05 \) mm were calculated. Shown in the figure are the off-axis distances \( x_{2} \) of the trajectories obtained at the exit plane. From this figure we see that while large off-axis aberrations occur for \( \theta \) greater than around ±40° or ±50°, good spatial resolution can be obtained by limiting the angle \( \theta \) to less than around ±30°. The lower panel of Fig. 11(a) shows the angles \( \theta_{2} \) of the trajectories at the exit plane. Fortunately, the shift in \( \theta_{2} \) caused by the electron-source size is small for any initial angle \( \theta \) from 0° to ±90° and it seems that it is possible to obtain good angular resolution over the full angle range in OPAL. Figures 11(b) and 11(c) are, respectively, the corresponding results to the OPALS in Figs. 10(a) and 10(b), showing no significant difference in the source size effect from the result shown in Fig. 11(a).

5. Discussion

On the basis of the cathode lens technique used in photoemission electron microscopy, we studied the possibilities of performing 2π-steradian photoelectron spectroscopy in a wide energy range. In usual PEEM and related instruments, 2π-steradian measurement is usually performed only for a low kinetic energy range up to some tens of eV, due to the limitation of the acceleration voltage applied to the cathode lens. For a fixed acceleration voltage, the spherical aberration of the cathode lens (defined for virtual rays) as well as the incidence angles to the objective lens considerably increases with increase of the initial electron kinetic energy. We showed that the spherical aberration of a cathode lens can be effectively reduced by deforming the cathode and the counter electrode. Here we considered the following combinations of the cathode and the counter electrode:

(a) a flat cathode and an open-convex counter electrode with a plane grid,
(b) a flat cathode and an open-convex counter electrode with no grid,
(c) a convex cathode and a plane grid counter electrode,
(d) a convex cathode and an open-convex counter electrode with a plane grid,
(e) a convex cathode and an open-convex counter electrode with no grid.

Comparing cases (a) and (b) and also cases (d) and (e), it was demonstrated that the use of a plane grid for the counter electrode is advantageous for reducing spherical aberration. The smallest spherical aberration is obtained for case (d). In this case, even a negative spherical aberration can be produced, while an ordinary cathode lens always has a positive spherical aberration. It should be noted that case (e), despite no use of a grid, has a significantly smaller spherical aberration than cases (a) and (c).

We next showed that the combination of a deformed cathode lens and a decelerating mesh lens provides a spherical aberration-corrected focusing lens with a full acceptance angle of ±90°. We express this lens as “OPAL”. We presented some basic designs of OPAL with different cathode lenses. If there is no spherical aberration in a cathode lens, a decelerating mesh lens with no spherical aberration can be used as is. On the other hand, if there is positive spherical aberration, as is almost the case for the considered cathode lenses, the decelerating mesh lens is designed with negative spherical aberration to cancel that positive spherical aberration. However, the cancellation over the full acceptance angle seems to be not so easy if there is a large positive spherical aberration in the cathode lens, and thus it would be desirable for OPAL to use a cathode lens with a small spherical aberration. Consequently, the OPAL designed with a cathode lens of case (d) has a considerably small spherical aberration over the full acceptance angle, as expected. However, this case has a disadvantage that the sample size must be small, limited by the cathode shape. A good alternative is to use a cathode lens of case (a). This case also allows good spherical aberration correction over the full acceptance angle.

OPAL can serve as the objective lens in various photoelectron spectrometers. Here an actual OPAL should be designed to achieve the best performance in an individual system. It is possible not only to design an own photoelectron spectrometer based on OPAL but also to combine OPAL with a commercial photoelectron spectrometer. The OPAL shown in this paper has a focusing angle of around ±11°. Thus, if the acceptance angle of a commercial photoelectron spectrometer combined with OPAL is less than ±11°, the focusing angle should be reduced by modifying the OPAL design or arranging an additional lens behind the OPAL. Combining OPAL and CHA in this manner, we can achieve a high energy resolution photoelectron spectrometer to measure full-range (±90°) 1D angular distribution at once. Moreover, it is possible to design an OPAL-CHA spectrometer to measure 2D angular distribution over a full half-sphere solid angle (2π steradian) at once, by arranging an energy-selecting slit (or aperture) and a projection lens at the exit of CHA.4–6

Here, we focus on the kinetic energy range in which OPAL can be applied with no limitation in its acceptance angle. In the OPAL shown in this paper, the acceleration voltage is set to 20 kV for electrons with kinetic energy of 1 keV. For other kinetic energies, all the voltages applied to OPAL including the acceleration voltage are changed in proportion to the kinetic energy in order to obtain the same focusing power. An actual OPAL can be designed so that it can use an acceleration voltage of at least up to 50 kV without discharge. Then, the maximum of the possible kinetic energy range reaches 2.5 keV. Atomic structure analyzes by photoelectron holography, atomic stereography, and diffraction spectroscopy are mostly performed in an X-ray region of less than around 1.5 keV. Thus, OPAL can perform well for those analyzes without decrease of the acceptance angle, while it seems to be difficult for usual PEEM and related instruments.

Fig. 8. (Color online) Omnidirectional photoelectron acceptance lens obtained by combination of a cathode lens in Fig. 3(a) and a decelerating mesh lens.
to achieve a full acceptance angle of ±90° or even a wide acceptance angle in that energy region.

Finally, we mention some remarks on the use of a plane grid and a mesh electrode. While the use of a plane grid in OPAL allows fine correction of spherical aberration over the full acceptance angle, it of course results in lower transmittance in OPAL than the case of not using it and can be disadvantageous for sensitivity of an analyzer. However, a high transmittance of around 80% or even 90% is possible in a plane grid, and using such a plane grid in OPAL, the decrease in transmittance can be suppressed to a small degree. We suppose that the mesh electrode has a transmittance of around 60%; then the expected transmittance of OPAL becomes around 50%. Another important remark is the disturbing effect of mesh (grid) holes,46,47) which can greatly degrade the image quality. To suppress this effect, the mesh and the grid holes are required to be as small as possible. Also important are the flatness of the plane grid and

Fig. 9. (Color online) (a) Combination of the simple cathode lens in Fig. 1(a) and a decelerating mesh lens and (b) combination of the cathode lens in Fig. 3(b) and a decelerating mesh lens.
the accuracy of the mesh electrode, which can greatly affect the focusing power of the OPAL. Summarizing the above, the plane grid and the mesh electrode should be high-accurate, fine-grained, and with high transmittance. This condition is relatively easy to achieve in the plane grid; however, the mesh electrode, having a curved shape, is not so easy to create under the condition. There are various methods to fabricate a curved mesh electrode, including a press molding method and an electroforming method. In any of the methods, a deeply curved mesh is difficult to fabricate under the above-mentioned condition; however, the concave shape of the mesh electrode of OPAL is much shallower than those of the mesh electrodes of the analyzers shown in Refs. 28–34. Thus, for OPAL, we expect that a mesh electrode can be better fabricated by a certain method.

6. Conclusion
Modifications of a simple cathode lens were studied in detail by ray-tracing calculations. We found that spherical
shown in Fig. 8, (b) OPAL shown in Fig. 10(a), and (c) OPAL shown in Fig. 10(b).

Fig. 11. Off-axis aberrations and final angles $\theta_f$ calculated in (a) OPAL shown in Fig. 8, (b) OPAL shown in Fig. 10(a), and (c) OPAL shown in Fig. 10(b).

aberration can be effectively reduced or corrected by using either or both of a convex cathode and a convex counter electrode, and that even negative spherical aberration can be produced in this way. We demonstrated that this approach allows a spherical aberration-corrected focusing lens with a full acceptance angle of $\pm 0.5^\circ$. The effect of photon beam size is expected to be negligible if the size is within 0.1 mm in diameter. An OPAL, designed with a deformed cathode lens and a decelerating mesh lens, would allow $2\pi$-steradian photoelectron spectroscopy in a wide energy range up to a few keV, which may open new horizons in photoelectron spectroscopy.

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