Application of atomic stereomicroscope to surface science

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Abstract

A stereograph of atomic arrangement was displayed directly on the screen of display-type spherical-mirror analyzer without any computer-aided conversion process for the first time. This stereoscopic photograph enables viewing three-dimensional atomic arrangement. This technique was realized taking advantage of the phenomenon of circular dichroism in photo-electron angular distribution. The azimuthal shifts of forward focusing peaks in a photo-electron angular distribution taken with left and right helicity light in a special arrangement are the same as the parallaxes in a stereo view of atoms. Hence a stereoscopic recognition of three-dimensional atomic arrangement is possible, when the left eye and the right eye respectively view the two images obtained by left and right helicity light simultaneously.

Keywords: Structure analysis; Photoelectron angular distribution; Circular dichroism; Stereo view; Stereomicroscope; Display-type spherical-mirror analyzer

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1. Introduction

Determination of three-dimensional atomic arrangement of surface is still a difficult problem because of the lack of periodicity in the surface normal direction. So far, have been developed many analysis methods, such as surface X-ray diffraction, X-ray standing wave method, low energy electron diffraction (LEED), reflection high energy electron diffraction (RHEED) and so on for this purpose. Among these methods the photoelectron holography [1–3], which is a recent extension of the photoelectron diffraction [4], has attracted much attention because it can in principle reconstruct the three-dimensional structure around a specific atom without the so called phase problem.

All methods mentioned above are diffraction methods, in which the obtained data are interference patterns in a reciprocal space and the analysis method is the Fourier transformation. On the other hand microscope can obtain direct structural image
without any conversion. The development of the STM (scanning tunneling microscope) has made it possible to obtain atomic images easily. However, STM cannot indicate the atomic arrangement because it only shows the corrugation of the surface electronic clouds, and cannot indicate the relation between the top atom and the second layer atoms. An electron microscope can produce atomic images. However, those images are two-dimensional projected images of three-dimensional lattice along the incident electron beam. Hence there has been no method to show a three-dimensional atomic arrangements directly. Recently, a new method of stereomicroscope of atomic arrangement was developed [5] by taking advantage of the phenomenon of circular dichroism in two-dimensional photoelectron angular distribution (2D-PEAD) [6].

In this article described are the analyzer used in the measurement, the characteristics of 2D-PEAD, and recent topics in circular dichroism in 2D-PEAD, such as “rotation of forward focusing peaks” [6], and “stereo microscopy of atomic arrangements” [5]. The forward focusing peaks in a 2D-PEAD pattern taken with left and right helicity light were found to shift azimuthally [6]. These shifts are the same as the parallaxes in a stereo view of atoms, and stereo photographs of atomic arrangement can be displayed directly on the analyzer without any computer-aided conversion process [5]. Stereoscopic recognition of three-dimensional atomic arrangement has become possible for the first time when the left eye and the right eye respectively view the two images obtained by left and right helicity light simultaneously.

2. Experimental method for two-dimensional photoelectron angular distribution

2.1. Conventional methods

2D-PEAD is extensively studied recently because it has full information in photoelectron spectroscopy, not only for structural analysis but also electronic state analysis such as Fermi surface mapping. 2D-PEAD can be obtained either by rotating sample [7], rotating analyzer, or by using display analyzer [8]. The relation between the orientation of the sample and the direction of the electric vector of photon changes in the rotating-sample method. The information obtained in the rotating-analyzer method and in the display analyzer methods is basically identical. In the case of rotating analyzer method, however, the measurement around the surface normal direction cannot be made with normal incident photons because the angular separation between the incident photon and detection direction is limited by the size of the analyzer. The 2D-PEAD around surface normal with normal incident photon is important to investigate the character of the bands [9].

Display-type spherical-mirror analyzer [10–12] can display 2D-PEAD at a specific kinetic energy at a time in a very wide acceptance angles without distortion. This advantage allows this analyzer to be used in a variety of applications including stereomicroscope. In a special arrangement, non-distorted stereo photographs are
directly displayed on its screen. Recently it was modified so as to achieve higher energy resolution and even wider acceptance angles [13,14].

2.2. Display-type spherical mirror analyzer

A display-type spherical mirror analyzer (DIANA) (Fig. 1) was invented in 1988 [10] and gradually improved [11,12]. Comparing with Eastman-type display analyzer [15,16], this analyzer has two advantages. One is that the angles of emission and detection of photoelectrons are exactly parallel; hence the pattern displayed on its screen is not distorted. The other is that the acceptance angle can be made infinitely wide, because the convergence to the exit aperture is exact irrespective of the angle and has no higher order terms. Hence one can measure the angular distribution of particles of one particular kinetic energy up to about $2\pi$ steradians at a time.

![Diagram of DIANA analyzer](image)

Fig. 1. New display-type spherical mirror analyzer (DIANA). G: hemispherical grid, Ob: obstacle rings on outer sphere Os, Gd: guard rings, S: sample, O: center, M: microchannel plates (MCP), P: phosphor screen, E: electron gun, D: deflector, SR: synchrotron radiation, PG: preretardation grids, and R: retardation grid. G is usually grounded and the electrons converge exactly to the exit aperture A with incident angles being parallel to their emission angles. The acceptance cone covered by the MCP is $\pm60^\circ$, and can be increased up to $\pm80^\circ$ by using the lens L.
The original analyzer has been applied to many surface studies, such as electron diffraction [17], two-dimensional photoelectron diffraction [18,19], photoelectron holography [20], electron stimulated desorption ion angular distribution (ESDIAD) [21–26], circularly polarized light photoelectron diffraction [6,27–29], two-dimen-
sional valence band analysis [30–39] etc. In the development of the analyzer, the
energy analysis method has been changed from “focus–defocus” [10] to “high-pass
and low-pass filter” [11,12]. The energy resolution of the conventional DIANA has
been about 1% of the pass energy.

Fig. 1 shows a schematic view of the new display-type spherical mirror analyzer
[13]. It consists of a hemispherical grid G, obstacle rings Ob and guard rings Gd. The
number of obstacle rings and guard rings are 260 and 20, respectively. The Ob’s are
axially symmetric with respect to the axis connecting the sample and the center O of
the hemisphere, and their inner surfaces are a part of concentric spheres. G is usually
grounded. The potential \( V(r) \) of Ob’s and guard rings are biased according to their
radii \( r \) from O by a formula \( V(r) = -2V_0(1 - a/r) \), where \( V_0 \) is the voltage corre-
sponding to the pass energy \( E_{\text{pass}} \) and \( a \) is the radius of the main grid G. The
electrostatic field in the space surrounded by the electrodes G, Ob and Gd is thus made
spherically symmetric with respect to the center O. The orbits of the emitted electrons
from the sample are subjected to this field after passing through G, and their loci are
ellipsoids obeying the Kepler’s law. Their trajectories inside G are straight lines.

The electrons converge exactly to the exit aperture A, which is located at the
symmetric position of S with respect to O. Their incident angles are exactly parallel
to their emission angles. Ob’s play essential roles in energy analysis by cutting un-
necessary electrons that have higher kinetic energies than the pass energy \( E_0 \) [11,12].
An electron having a little higher energy flies along a little outside locus and hit into
the electrodes and absorbed or scattered, and hardly passes through the small ap-
erture A. Thus the Ob’s work as a “low-pass filter”. On the other hand, retarding
grid R works as a “high-pass filter” which retards the electrons having lower kinetic
energies than the applied potential.

The electrons having passed through the retarding grid are amplified by a pair of
microchannel plates (MCP) M and converted by a phosphor screen P to light pulses,
which are detected from outside the vacuum chamber by a cooled CCD camera. The
2DAD on the screen is the same as the original angular distribution in front of the
sample. An electron gun E is mounted for the purpose of LEED measurement.
Synchrotron radiation SR is introduced through a hole. The acceptance cone
covered by the MCP is \( \pm 60^\circ \). This acceptance angle can be increased up to \( \pm 80^\circ \) by
using the lens L. The diameter of the main grid was two-times enlarged from 150 mm
of the old analyzer to 300 mm. The shaping accuracy of Ob was intended to be
within 50 \( \mu \text{m} \). The energy resolution \( (\Delta E/E_{\text{pass}}) \) of the present analyzer has been
estimated to be 0.25% of the pass energy, which is four times higher than before.
Angular resolution was estimated to be 0.6°.

The problem of it is that the size is very large (about 700 mm) and fabrication
method is difficult (ceramic plating and making contact with evaporated thin layer of
Au) and a miscontact sometimes happened. More recently the size was reduced half
and the reliability was improved with keeping the performance the same [14]. Fig. 2
shows the outer sphere of the newest analyzer made of insulator so that the mis-
contact cannot occur.

3. Two-dimensional photoelectron angular distribution and holography

Fig. 3 shows various features in 2D-PEAD. The atom O produces a photoelectron wave $\psi^0$ when an X-ray excites it. $\psi^0$ produces a scattered wave $\psi^S$ when it hits a scatterer atom. The 2D-PEAD is described by a square of the sum of $\psi^0$ and all $\psi^S$s from nearby atoms. When the emitter atom is in the surface region, the characteristic features are “forward focusing peaks” and “interference rings”. When the emitter atom is in the bulk they produce “Kikuchi bands” and “Kossel lines”.

Recently, many methods for three-dimensional analysis using 2D-PEAD patterns have been developed, such as photoelectron holography [1,2,20], Kikuchi-electron holography [3,40,41] or correlated thermal diffuse scattering [42]. Holography analysis produces the three-dimensional atomic arrangement around the emitter atom by a Fourier transformation of the 2D-PEAD. Photoelectron holography is more powerful than Kikuchi-electron holography because it can specify an emitter by selecting the kinetic energy of photoelectrons. These holographic analyses are rather direct method because they do not need a trial model of atomic arrangement. Although its accuracy is not high (about 0.2 Å), it is useful for a distinction of various models or for making a first trial model for LEED analysis.

3.1. Surface photoelectron diffraction

When the kinetic energy of a photoelectron is higher than several hundred eV, a strong forward focusing peak appears along the direction connecting the emitter and the scatterer. Fig. 4 is an example of forward focusing peaks. It is a 2D-PEAD of the
Fig. 3. Various features of two-dimensional photoelectron angular distribution (2D-PEAD).

Fig. 4. 2D-PEAD of W 4f photoelectrons from the first-layer W atoms on W(110)1 × 12-O surface.
W 4f photoelectrons, from only the first-layer W atoms on W(110)1 × 12-O surface [43]. The photoelectrons from the first-layer W atoms are easily distinguished from those of bulk W atoms utilizing a large chemical shift (0.73 eV). Fig. 4 is plotted by stereographic projection method. The center of the pattern corresponds to the surface normal direction and the edge corresponds to the polar angle of 90°. The excitation light is Al Kα.

We can see six strong forward focusing peaks. There are two domains on this surface, where O atoms are sitting at the middle of upward and downward triangles, as shown in Fig. 5. The directions of the peaks A, B, C, A', B', and C' correspond to the direction of oxygen atoms seen from the first-layer W atoms.

The measurement of the directions of forward focusing peaks is sufficient for three-dimensional analysis when the structure is simple. In this case the position of adsorbed oxygen was determined directly from the directions of these peaks [43]. Usually, however, the precise structure can be obtained by comparing the observed pattern with the pattern calculated including multiple-scattering.

3.2. Bulk photoelectron diffraction and Kikuchi pattern

Fig. 6 shows an example of bulk photoelectron diffraction pattern. We can see strong peaks in the [100] and [111] directions, which are forward focusing peaks along these crystallographic axes. The broad lines connecting these axes are called Kikuchi bands. The band edge is dark here, but it will be bright if the mean-free-path of photoelectrons become longer at higher kinetic energies. They are called Kikuchi lines or Kossel lines. Original spherical photoelectron waves are considered as plane waves in a distance. The Bragg reflections of these plane waves produce these Ki-

![Fig. 5. Top view of W(110)1 × 12-O surface.](image-url)
kuchi patterns. We can know the crystal orientation and the interlayer spacing easily from Kikuchi patterns.

3.3. Photoelectron holography

3.3.1. Interference rings

The interference rings are hardly seen in Fig. 4, because the forward focusing peaks are much stronger than them. Fig. 7(a) shows an example of such rings. This pattern is the angular distribution of O Is photoelectrons from adsorbed oxygen on W(110)1 × 12-O surface. We can see first, second, and even third order rings around the axes \(\langle 1\bar{1}1\rangle\), \(\langle 001\rangle\), and \(\langle 1\bar{1}0\rangle\). Because the O atoms are sitting at the top of the surface, the forward focusing peaks must appear along the surface, which are out of the measurement area. Hence, we can see only diffraction rings. The fact that we can see these rings, whose center is at the polar angle of 90°, means that O atoms are sitting at the top of the surface. The order of the size of the ring is \(\langle 1\bar{1}1\rangle\), \(\langle 001\rangle\), and \(\langle 1\bar{1}0\rangle\), which means that the nearest atoms are in \(\langle 1\bar{1}1\rangle\) directions, and the second and the third nearest atoms are in \(\langle 001\rangle\) and \(\langle 1\bar{1}0\rangle\) directions, as shown in Fig. 5.

Fig. 8 is a diagram to show how these rings appear. Neglecting the phase of the scattering factor of atom A, the phase difference between \(\Psi^0\) and \(\Psi^S\) is \(kR - kR \cos \theta\). Hence the rings appear at \(\theta\) satisfying

\[
kR(1 - \cos \theta) = 2\pi n,
\]

when \(n\) is an integer. In Fig. 7(b) the data of Fig. 7(a) is plotted so that the distance from the center is in proportion to \(\cos \theta\), which is also proportional to the lateral
momentum of the photoelectron. We can see that the rings in Fig. 7(a) became parallel lines in Fig. 7(b). This fact implies that Eq. (1) holds. The interatomic distance $R$ can be calculated easily from the spacing $\delta(k \cos \theta)$ as $R = 2\pi/\delta(k \cos \theta)$. The photoelectron holography utilizes these interference rings that appearing in many directions.
3.3.2. Photoelectron holography

It has been recognized that the holographic method (Fourier transformation) to obtain the real structure is not so straightforward. The difficulty in the photoelectron holography comes mainly from the strong forward focusing peaks in the 2DAD pattern, which always appear when the emitter atom is below the scatterer atoms. Many efforts have been made to eliminate the disturbance from these forward focusing peaks by means of SWIFT [3], SWEEP [44], energy extension [45], or differentiation [46].

Because the pattern of Fig. 7 consists of only interference pattern, it seems a good pattern for the holographic analysis. The holographic reconstructed structure image of Fig. 7 is shown in Fig. 9 [47]. We can see strong peaks near the expected positions indicated by circles. The accuracy, however, is about 20% in this case.

4. Circular dichroism in two-dimensional photoelectron angular distribution

4.1. Rotation of forward focusing peaks

In general, a forward focusing peak has information about the direction from the emitter to the scatterer but does not have the information about the distance between them. Recently, it was found that the forward-focusing peak-positions in a 2D-PEAD excited by a circularly polarized light rotate in the same direction as the rotation of the electric vector of the light [6,48].
4.1.1. Si(001)

The first experiment on circular dichroism in photoelectron diffraction was made on Si(001) [6]. Fig. 10(a) and (b) show the 2D-PEAD of Si 2p photoelectrons at a kinetic energy of 250 eV. The circularly polarized photons are incident normal to these figures and the rotation direction of its electric vector is (a) counter clockwise ccw and (b) clockwise cw, respectively. The crosses show the calculated (the formula is described afterward) peak positions for ⟨110⟩, ⟨112⟩, ⟨113⟩, and ⟨115⟩ forward peaks from outside to inside, respectively. These forward peaks must lie in symmetric planes, which are shown by vertical and horizontal white lines in the figure, if the excitation light were linearly- or non-polarized light. We can see all peaks rotate to the same rotational direction with that of excitation circularly polarized light in rather good agreement with the predicted directions shown by crosses. This is a strong circular dichroism in 2D-PEAD (we call it as CDPEAD hereafter) for non-chiral and non-magnetic materials.

4.1.2. W(110)1×1-O

Fig. 11(a) and (b) show another examples of CDPEAD for W(110)1×1-O [48]. The six peaks are the same forward focusing peaks seen in XPD (Fig. 4). Here the photoelectron kinetic energy is 317 eV, the photon was incident normal to the surface, and the polar take-off angle range is from 61.0° to 73.5°.

Fig. 11(a) is the result of linearly polarized (LP) and (b) is of left circularly polarized (LCP) light. Here the definition of LCP is that the rotation direction of the electric vector of the photon in the plane of Fig. 11(b) is counter-clockwise (ccw). Because the emitter atom is the top layer W atom and there are only O atoms above

![Fig. 10. 2D-PEAD of Si 2p photoelectrons from Si(001) surface at 250 eV measured with (a) counter-clockwise and (b) clockwise circularly polarized light.](image-url)
Fig. 11. 2D-PEAD of W 4f photoelectrons from the first-layer W atoms on W(110)1 × 12-O surface measured with (a) linearly polarized and (b) counterclockwise circularly polarized light.

it, these forward peaks in Fig. 11 are considered to be dominated by single scattering. It is clear that there is a tendency for the pattern to shift counterclockwise with LCP excitation in Fig. 11(b).

4.1.3. Simple formula for the rotation angle

All these shifts are considered to be rotation of the forward focusing peaks with respect to the photon incident direction. This rotation has been explained as being due to the transfer of the angular momentum of photons to the photoelectrons, whose z component ratio has been biased while being excited by the circularly polarized light.

The rotation angle \( \Delta \) of the peak around the photon incident axis is reproduced well by the simple formula [6]

\[
\Delta = \tan^{-1} \frac{m}{kR \sin^2 \theta} \cong \frac{m}{kR \sin^2 \theta}
\]  

(2)

Detailed theoretical works [27,49,50] support this formula. Here, \( m \) is the magnetic quantum number (z component angular momentum) of the photoelectron, \( k \) is its wave number, \( R \) is the internuclear distance between the emitter and the scatterer, and \( \theta \) is the angle between the photon incident direction and the outgoing photoelectron direction, as shown in Fig. 12.

When the position vector of the scatterer from the emitter is described as \( (R, \theta, \phi) \), the peak position observed by using cw and ccw circularly polarized light appears at \( (\theta, \phi \pm \Delta) \). Using this formula we can construct three-dimensional atomic structure directly by measuring the azimuthal shift of the forward focusing peaks in the photoelectron diffraction patterns excited by the circularly polarized light [28,29].
Hence, this method positively utilizes the forward focusing peaks for structural analysis.

4.2. Theoretical analysis

4.2.1. Rotation angle and the orbital angular momentum

The characteristic feature of the photoelectrons excited by circularly-polarized light is that it has an orbital angular momentum around the incident light. We consider the photoelectrons from an initial core state $\Psi_i$ with the quantum number of the angular momentum $l'$ and its $z$-component $m'$. Its angular part can be described by a spherical harmonic $Y_{l'm'}(\theta, \phi)$. The quantum axis ($z$-axis) is the traveling direction of the incident circularly polarized light.

The transition operator $r \cdot e$ in photoexcitation process within the dipole approximation is conveniently expressed using the spherical harmonics.

$$ r \cdot e = \sum_{\mu = -1}^{1} \sqrt{\frac{4\pi}{3}} e_\mu Y_1^\mu(\theta, \phi) r $$

where $e$ is the electric vector of the photon. The transition operator for a circularly polarized light with the angular momentum of photon $\sigma_{\mu}(= \mu)$ is expressed by the $\mu = \pm 1$ term. The transition matrix to the final state wave function $\Psi_f$ with the quantum number $l$ and $m$ is written as [51]

$$ \langle \Psi_f | r \cdot e_{\pm} | \Psi_i \rangle = \sum_{l = \pm 1} R_{nm \rightarrow kl} c^1(lm, lm') $$

(4)
where $R_{nlm ightarrow kl}$ is a radial matrix element, and $c^{1}(lm, l'm')$ is the Gaunt coefficient. This $c^{1}(lm, l'm')$ is not zero only when $m = m' \pm 1$. Hence, a circularly polarized light with the angular momentum of photon $\sigma_{hv}$ of $\pm 1$ excites the ground state with the magnetic quantum number $m'$ to the final state with $m = m' \pm 1$. In other words, the angular momentum that the electron gains by photoexcitation is $\pm 1$.

First we consider a final state with only one $m$ component. The final state wavefunction outside the emitting atom is expressed as

$$\Psi \propto \frac{e^{ikr}}{r} Y_{lm}(\theta, \phi) \propto \frac{e^{ikr}}{r} \Theta_{lm}(\theta)e^{im\phi}$$  \hspace{1cm} (5)$$

where $\Theta_{lm}(\theta)$ is the $\theta$-function in the spherical harmonic.

The propagation direction of this final state wavefunction is intuitively calculated considering the wave front of Eq. (5). The phase of the wave is expressed as

$$\exp[i(kr + m\phi)]$$  \hspace{1cm} (6)$$

The equation of the constant phase surface is expressed as

$$r = -\frac{m\phi}{k} + C$$  \hspace{1cm} (7)$$

where $C$ is a constant. This curve is a spiral, and is shown in Fig. 13 by solid and broken lines. The wave propagates perpendicular to these lines, which is the wave front, as shown by the thick arrow in Fig. 13. The shift of the forward direction $\Delta$ from the OA direction is expressed as

$$\Delta = \tan^{-1}\frac{m}{kR \sin^2 \theta} \propto \frac{m}{kR \sin^2 \theta}$$  \hspace{1cm} (8)$$

The physical base of the azimuthal shift is summarized as follows. The forward focusing peak appears because the phases of all wavelets on the wave front being scattered by each infinitesimal atomic potential coincide in the forward direction. This “forward direction” is perpendicular to the wave front of the wave. When the wave has angular momentum, the wave front hit the scatterer at an angle. Hence the forward direction of the photoelectron wavefunction is inclined at the scatterer. In other words, the peak does not shift from the forward direction but the forward direction has shifted already before the scattering.

This formula for the direction of the forward focusing peak is exact when the photoelectron wavefunction can be expressed by only one component of angular momentum. This condition is satisfied in the excitation of the $s$ core, where final $m$ should be $\pm 1$. In the following, described is more general case of excitation.

The transition probability to the $l' + 1$ final state is much higher than that to the $l' - 1$ final state. In the case of the photoexcitation from the Si 2p ($l' = 1$) core state by a photon of $\sigma_{hv} = +1$, which is the case of Ref. [6], the final $l$ is either 2 or 0, and the final $(l, m) = (2, 2), (2, 1), (2, 0)$ and $(0, 0)$. The probability of realizing these three

\footnote{$c^{1}(lm, l'm') = \frac{1}{\sqrt{(2k+1)}} \int Y_{lm}(\theta, \phi) Y_{l'm'}(\theta, \phi) \sin \theta d\theta d\phi$.}
Fig. 13. Constant phase surface of photoelectron wave with angular momentum $m$.

$l = 2$ final states is much higher than that for $l = 0$, being about 13:1 at the photon energy of around 350 eV [51].

The probabilities of realizing these three final $m$ values are not equal because the Gaunt coefficients are different. For example, the probabilities for $(l, m) = (2, 2) : (2, 1) : (2, 0)$ are (6:3:1). When $\sigma_{iv}$ is $-1$, the ratio for $m = (-2, -1, 0)$ is also (6:3:1). Even when the spin-orbit interaction of the core state is considered, this ratio is unchanged for each spin-orbit split component $P_{1/2}$ and $P_{3/2}$. This ratio, however, is that of the total cross section over $4\pi$ steradian and it depends on the emission angle $\theta$ (but does not depend on $\phi$).

4.2.2. Effective $m$ value

The effective $m$ value, which is an average of possible final-state $m$ values at certain polar angle, is discussed in detail for the analysis. The effective $m$ as a function of $\theta$, $m^*(\theta)$, has been derived considering the spin-orbit interaction [29]. When the small contribution of $l' - 1$ component is neglected, the $m^*(\theta)$ for general $l'$ initial state is expressed as

$$m^*(\theta) = \frac{\sum_{m'=-\mu}^{\mu} m|c_{l'+1,m}^{l'}(l', m, l', m')\Theta_{l'+1,m}|^2}{\sum_{m'=-\mu}^{\mu} |c_{l'+1,m}^{l'}(l', m, l', m')\Theta_{l'+1,m}|^2}, \quad (9)$$

where $m = m' + 1$ for $\sigma_{iv} = +1$. More general formula is written in [29].

The angular dependence of the contribution of each $m$ state in the final state, which is $|c_{l'+1,m}^{l'}(l', m, l', m')\Theta_{l'+1,m}|^2$, is shown in Fig. 14 for $l' = 3$. The effective $m$ value $m^*(\theta)$ of Eq. (9) is also shown in Fig. 14. The neglect of $l - 1$ channel causes an error of a few percent. Eq. (9) is convenient because it depends only on the initial angular momentum $l'$ and does not depend on energy or atomic species. The applicability of this analysis was discussed in a typical example of W(1 1 0) surface [28].
5. Stereograph by rotation of forward focusing peaks

5.1. Stereoscopic photographs

A stereoscopic photograph consists of a pair of photographs; one is that to be seen by the left eye, and the other is to be seen by the right eye. The positions of an identical object in the two photographs differ by an amount parallax, which is inversely proportional to the distance from the observer. Assume that you are facing the x direction and looking at an object A with your right and left eyes at E and F, respectively, as shown in Fig. 15. When the position of the object A is described as $(R, \theta, 0)$ in polar coordinates, the relation between the distance $R$ and the azimuthal shift $\Delta$ of the object A in the two photographs is described as

$$\Delta = \tan^{-1} \frac{b}{R \sin \theta}, \quad (10)$$

where $b$ is one half of the distance between the two eyes (OE and OF in Fig. 15).

5.2. Stereo photograph of atomic arrangement

Two photographs of atomic arrangement satisfying the condition of Eq. (10) can be obtained by using a display-type spherical-mirror analyzer described above in the measurement of circular dichroism in 2D-PEAD (CDAD). For example, the CDAD pattern of W 4f photoelectrons from the W(1 1 0) surface at a kinetic energy of 800 eV is shown in Fig. 16(a) and (b) [5]. These patterns are observed on a flat screen of
Fig. 15. Stereo view of atom A with right and left eyes at E and F, respectively.

Fig. 16. Stereo photograph of W crystal. If we view (a) with the left eye and (b) with the right eye, we can produce an image of a three-dimensional arrangement of atoms in W(1 1 0) crystal as shown in Fig. 17(a).

the analyzer. The angular range on the screen is about $\pm 60^\circ$. Fig. 16(a) and (b) were taken with cw and ccw circularly polarized light, respectively.
The center of the figure nearly corresponds to the surface normal direction, which is the [1 1 0] axis, and the horizontal direction is [0 0 1]. The ccw and cw excitation light is incident 45° inclined to the surface normal, which is the [0 1 0] direction, as shown in Fig. 17(b). We can see five forward focusing peaks, such as A, B, C, in the directions shown in Fig. 17(a). These forward peaks are produced by nearest neighbor atoms A, B, and C in Fig. 17(b). In XPD [43], the positions of these forward peaks are the same as these directions, but their positions in Fig. 16(a) and (b) are slightly different from them. The patterns obtained by cw and ccw light are symmetric with each other with respect to the central vertical line. For example, the [1 0 0] peak A is slightly off the center to the right in (a) and left in (b), as shown by dots in the figure.

Because in both cases, (Eqs. (8) and (10)) the angular shift \( \alpha \) of the object is inversely proportional to the interatomic distance \( R \) from the emitter, these patterns can be considered as stereoscopic photographs. The necessary condition is that the direction of the photon be parallel to the z-axis in stereoscopic photography. Then, if we view the clockwise (cw) circularly polarized-light photoelectron angular distribution pattern (Fig. 16(a)) with the left eye and the ccw pattern (Fig. 16(b)) with the right eye, we can image a three-dimensional arrangement of atoms in W(1 1 0) crystal as shown in Fig. 17(a). The A atom looks closer, the B and C atoms look farther, and others look the farthest. Moreover, near atoms look bigger and far atoms look smaller as in the case of real view. The elongation of atom A is due to the projection of the pattern to a flat screen.

To recognize the three-dimensional arrangement properly, it is necessary that the fields of vision of both images should be the same as that in the measurement. The solid angle of these pictures is about \( \pm 60^\circ \) in the measurement whereas it is only \( \pm 15^\circ \) when these pictures are viewed by using usual stereo-viewer. Special projection method, such as a combination of polarized light projection and polarized glasses for individual image, is necessary to realize the field of vision \( \pm 60^\circ \). If it is realized, the

![Figure 17](image_url)

Fig. 17. (a) Three-dimensional configuration of atoms imaged by stereo photograph of Fig. 16. (b) Structure of W crystal.
oval of atom A can be seen as a circle, and the inter-atomic relation is to be proportional to the real relation.

When Eqs. (8) and (10) are identical, there is no distortion. The necessary condition is that \(m/(k \sin \theta)\) be constant in Eq. (8). For the objects in the \(x-y\) plane, there is no distortion because \(m/(k \sin \theta)\) is constant. In this case, the magnification ratio is \(bk/m\). When \(b = 3\) cm, \(k\) is 14 Å\(^{-1}\) (for the kinetic energy of 800 eV), and \(m = 4\), the magnification ratio is about \(2 \times 10^{10}\), which is 2000 times higher than usual electron microscope. When \(\theta\) is not 90°, the sine function in the denominator of Eq. (8) would make the atoms close to the viewer. However, this distortion is hardly present in the actual case because the value \(m\) in Eq. (8) is not constant, and the \(\theta\) dependence of \(m^*(\theta)\) is close to \(\sin \theta\) [21], as shown in Fig. 12.

In this method the forward focusing peaks should be clearly seen in the 2DAD pattern. This restriction implies that the emitter atom must lie under the scatterer atoms and that the kinetic energy of the photoelectron should be above several hundred eV. The photoelectron diffraction and the multiple-scattering effect could modify the forward focusing peaks, but these effects are order of magnitude smaller than the forward peaks. The peak positions have been reproduced well so far not only for the adsorbate single-scattering case [48] but also for the bulk multiple-scattering case when the forward focusing peaks are clearly seen [28]. Hence, this analysis is applicable not only to the molecules adsorbed on surfaces but also to crystalline substances.

5.3. Stereomicroscope

This stereoscopic photograph can be obtained directly on the screen of above mentioned display-type spherical-mirror analyzer [10–14] without any computer-aided conversion process. Rotatable analyzer method to measure photoelectrons two-dimensionally can also take this stereoscopic photograph with the aid of computer. However, rotating-sample method produces much-distorted image. Other types of display analyzers such as Eastman-type analyzer [15,16] can also be used to take stereoscopic photographs. In this case, however, the pattern obtained on the screen is distorted, and computer processing is inevitably necessary to make the photograph. Because only the display-type spherical-mirror analyzer can obtain a distortion-free image in much wider solid angles than the Eastman-type one, it is the best analyzer to take stereoscopic photographs.

Real-time observation will help in understanding the dynamics of atoms. The problem in all atomic-structure analysis methods so far (electron diffraction, X-ray diffraction, photoelectron diffraction and holography) is that the data obtained are that of reciprocal space, and a real-space structure cannot be imaged before conversion. It takes some time to calculate the atomic structure, and these methods are thus not suitable for real-time analysis. The situation is even worse for fluorescent X-ray holography [52] because the time for data collection is much longer than that for electron emission holography.

Because the stereo photographs are presented directly on the screen, real time observation is possible. When the time required to switch the helicity of circularly
polarized light is reduced and the signal intensity increases, this display analyzer will give an opportunity for real-time stereoscopic observation. On the screen of DIANA the stereo photographs for right eye and left eye will be displayed alternately by switching the helicity of the light. Special glasses can transmit each image to each eye by switching the transmittance of each glass with the same period. In this way one can image three-dimensional atomic arrangements as shown in Fig. 18 in real time. Although it takes about 30 min at present to obtain a photograph, the development of synchrotron radiation and measurement techniques will continue to shorten the measurement time. In the near future, measurement times at least as short as that of video rate "real time" will be achieved.

Because this technique uses photoelectrons, the photographs become element- and chemical-state-selective by selecting the kinetic energy of photoelectrons. Because this method is a microscope, single molecule could be viewed in principle. For the observation of single molecule the light should be focused to a small region so that the background is suppressed. If we could hold a molecule on a sharp needle such as STM tip the observation would be possible in future.

References