Reconstruction Algorithm for Atomic Resolution Holography∗

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Atomic resolution holography, for example photoelectron holography and x-ray fluorescence holography, is a type of atomic structural analysis. These measurement methods can visualize the three-dimensional atomic structure around the target atomic site. These methods can also measure the atomic structure around an impurity in the crystal or an adsorbate on the crystal. The atomic image is obtained from the measured hologram using a reconstruction calculation. We developed scattering pattern extraction algorithm with maximum entropy method (SPEA-MEM) for atomic image reconstruction. In this paper, we describe the theory of the SPEA-MEM.

Keywords: Photoelectron diffraction measurement; Photoelectron holography; Soft X-ray photoelectron spectroscopy; X-ray scattering, diffraction, and reflection

I. INTRODUCTION

Recently, atomic resolution holography measurements, such as photoelectron holography [1], Auger-electron holography [2, 3], an inverse mode of photoelectron holography [4], x-ray fluorescence holography [1] and an inverse mode of x-ray fluorescence holography [5] were dramatically developed, because these atomic resolution holography have unique features. These methods can visualize a three-dimensional (3D) local atomic structure around a target atomic site for not only bulk but also impurity in crystal and adsorbate on crystal is observable. In order to see the 3D atomic image from the measured hologram, a reconstruction calculation is carried out. The algorithm for the reconstruction calculation is a key point to see the atomic image.

Barton proposed atomic image reconstruction algorithm based on Fourier transform [6, 7]. This algorithm requires multi-energy hologram. This algorithm is effective for the inverse fluorescence holography. In the case of the normal mode of the x-ray fluorescence holography, it is difficult to see the atomic image using Barton algorithm because it is difficult to measure the multi-energy hologram. In the case of the photoelectron holography (also the Auger electron holography and the inverse mode of photoelectron holography), Barton algorithm cannot reconstruct atomic image because phase shift effect in the photoelectron scattering process is serious. Recently, we developed a reconstruction algorithm SPEA-MEM (scattering pattern extraction algorithm with maximum entropy method) [8–11] not based on the Fourier transform. This algorithm can reconstruct the atomic image from not only a single-energy photoelectron hologram but also a single-energy fluorescence hologram. In this paper, we describe the SPEA-MEM.

II. RECORDING PROCESS AND SIMULATION

At first, we describe a recording process of the photoelectron holography. A schematic view of the recording process is shown in Fig. 1. (1) Excitation light excites a

![FIG. 1: Schematic view of photoelectron holography.](http://www.sssj.org/ejssnt)
an observed L_3VV Auger electron hologram of Cu(001). (b) A simulated Auger electron hologram. The kinetic energy is 914 eV. The emitted waves of f angular momentum and spherical cluster of 241 atoms are utilized.

(1) The emitted wave function and kinetic energy were set to be f wave and 914 eV, respectively. A spherical cluster of 241 atoms was utilized.

The simulated result for the Cu(001) Auger electron hologram is shown in Fig. 2(b). The emitted wave function and kinetic energy were set to be f wave and 914 eV, respectively. A spherical cluster of 241 atoms was utilized. Figure 2(a) is the experimental result [9] measured at SPring-8 BL25SU using two-dimensional display analyzer [14]. Simulated result exhibits good agreement with the experimental result.

(a) (b)

The phase shift effect is exactly taken into account. The algorithms can visualize the 3D atomic image from a single-energy photoelectron hologram.

We defined a scattering pattern function as

\[ t(k, a) = \sum_{L} 2\text{Re} [\varphi_L(k)\psi_L(k, a)] + |\psi_L(k, a)|^2. \]  

The hologram function can be written as

\[ \chi(k) = \sum_h t(k, a_h). \]

### III. RECONSTRUCTION

Barton proposed the algorithm to reconstruct the 3D atomic image using Fourier transform. The algorithm required the multi-energy hologram using the photoelectrons with a number of the kinetic energies. Therefore, the energy tunable light source and long measurement time were required. However, this algorithm cannot give a clear atomic image. Some improved reconstruction algorithms based on the Fourier transform were proposed. However, they did not always give a clear atomic image because the phase shift problem caused by an electron scattering process at a scatterer atom is serious. Recently, we developed some advanced algorithms to reconstruct the 3D atomic image. In the algorithm, the phase shift effect is exactly taken into account. The algorithms can visualize the 3D atomic image from a single-energy photoelectron hologram.

We defined a scattering pattern function as

\[ t(k, a) = |a| \sum_{L} 2\text{Re} [\varphi_L * (k)\psi_L(k, a)] + |\psi_L(k, a)|^2. \]  

The hologram function can be written as

\[ \chi(k) = \sum_h t(k, a_h). \]
The scattering pattern function for Cu is shown in Fig. 3. The angular momentum of the emitted Auger electron was set to be \( f \). The Forward focusing peak (FFP) appears on the \( z \)-axis. Some rings around the \( z \)-axis also appear. (b) The scatterer is not located on the \( z \)-axis. The atomic distance is 2.55 ˚A. (c) The scatterer is located on \( z = 3.61 \) ˚A. The scattering pattern function for Cu is shown in Fig. 3. The angular momentum of the emitted Auger electron was set to be \( f \). The Forward focusing peak and center of the interference rings indicate the direction of the scatterer atom as shown in Fig. 3(b). The spatial frequency of the interference rings implies the atomic distance between the emitter atom and scatterer atom as shown in Fig. 3(c).

The hologram function can be described as the sum of the scattering pattern functions. Then, we extended the equation as

\[
\chi(k) = \int g(a)/|k,a| da. \tag{5}
\]

Here, the three-dimensional atomic distribution function \( g(a) \) is introduced. When the function is defined as

\[
g(a) = \sum_h \delta(a - a_h) |a|, \tag{6}
\]

Eqs. (4) and (5) are equivalent.

In order to obtain the atomic distribution function \( g(a) \) from the hologram function \( \chi(k) \), we used a voxel with \( N \times N \times N \) mesh. The number of voxel is \( N^3 \). On the other hand, the number of the pixel of the hologram is given by the angular mesh \( M^2 \). In the case of the single-energy hologram, the number of the unknown parameters \( N^3 \) greater than that of known parameters \( M^2 \). Therefore, it is difficult to solve this equation.

We found that the voxel can be estimated by the maximum entropy method. We constructed an algorithm called SPEA-MEM (scattering pattern extraction algorithm with maximum entropy method). The entropy is defined as

\[
S = - \sum_j g_j^{(n)} \ln \frac{g_j^{(n)}}{g_j^{(n-1)}} - \lambda C, \tag{7}
\]

\[
C = \frac{1}{N} \sum_i \left| \frac{\chi^{exp}(k_i) - \chi^{cal}(k_i)}{\sigma_i} \right|^2 - 1. \tag{8}
\]

When the entropy is maximized the atomic distribution function is obtained. This calculation is iteratively applied. The schematic view of the iteration is shown in Fig. 4. The iteration is carried out as follows; (1) An electron hologram \( \chi^{exp}(k) \) is given. Initial values are given to a calculated hologram \( \chi^{cal}(k) \) and a voxel \( g(a) \). (2) Residual error between \( \chi^{exp}(k) \) and \( \chi^{cal}(k) \) is obtained. (3) The residual error is converted to a real-space residual error using scattering pattern matrix. (4) The voxel values are corrected by the real-space residual error using the maximum entropy method. (5) A data processing for voxel value is applied. (6) The voxel is converted to the calculated hologram \( \chi^{cal}(k) \) using scattering pattern matrix. (7) Jump to (2). This sequence iterates until convergence of voxel values.

In the case of a normal SPEA-MEM algorithm, no data processing for voxel value is applied. We applied the normal algorithm to the Cu(001) Auger electron hologram. The result is shown in Fig. 5. We succeeded in reconstructing a clear atomic image. In order to obtain clearer atomic image, there is some approach. (a) Use of multi-energy electron hologram. (b) Use of translational symmetry information. Especially the approach (b) is simple.
We applied the translational symmetry operation at (5) the data processing for voxel. The results are shown in Fig. 6. Very clear atomic image was reconstructed.

Finally, we mention about the x-ray fluorescence holography (XFH). In the case of the inverse fluorescence x-ray

FIG. 5: Reconstructed real space images using an experimentally-derived Cu(001) Auger electron hologram (Fig. 2(a)).

FIG. 6: Reconstructed real space images from an experimentally-derived PEH of Cu(001). The kinetic energy of photoelectron is 818 eV. The SPEA-MEM with the translational mixing operation was utilized.

FIG. 7: The scattering pattern functions for IXFH with photon energy of 12 keV. (a) The Au scatterer located at \( z = 2.88 \) Å. Some rings appear around the \( z \)-axis. (b) The Au scatterer located at \( z = 4.08 \) Å.

FIG. 8: An IXFH of Au(001). The photon energy is 12 keV.

FIG. 9: Reconstructed real space images from a single-energy IXFH of Au(001) (Fig. 8). The SPEA-MEM with the translational mixing operation was utilized.
holography (IXFH), it is possible to measure the multi-
energy hologram. The phase shift effect of the IXFH
is not serious. Therefore, Barton algorithm is effective.
However, many holograms recorded by changing photon
energy are required. In the use of the SPEA-MEM, it
will be possible to reduce the number of the holograms.
The scattering pattern function for the IXFH is different
from that for the photoelectron holography. A sample for
the scattering pattern function for hologram is shown in
Fig. 7. We applied the SPEA-MEM for the single-energy
IXFH of Au crystal (Fig. 8) using translational symmetry.
The results are shown in Fig. 9. Clear Au atomic image
was reconstructed. The SPEA-MEM is effective even for
the IXFH and XFH.

IV. SUMMARY

In summary, we have developed a simulation theory for
the photoelectron holography, and a reconstruction the-
ory SPEA-MEM. This method gives clear 3D atomic from
a single-energy photoelectron hologram (Auger electron
hologram and inverse mode of photoelectron hologram).
The SPEA-MEM can also reconstruct the atomic image
from x-ray fluorescence holography.

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