Optimization of Incident Electron Energy for Internal-Detector Electron Holography with Monte Carlo Simulation

Akio Uesaka†
Tohoku Techno Arch CO., LTD, 2-1-1 Katahira, Aoba-ku, Sendai 980-8577, Japan and
HORIBA, Ltd., 2 Miyanohigashi, Koshoin, Minami-ku, Kyoto 601-8510, Japan

Kouichi Hayashi
Institute for Materials Research, Tohoku University,
2-1-1 Katahira, Aoba-ku, Sendai 980-8577, Japan

Tomohiro Matsushita
Japan Synchrotron Radiation Research Institute (JASRI),
SPRING-8, 1-1-1 Kouto, Sayo, Hyogo 679-5198, Japan

Shigetoshi Arai
HORIBA, Ltd., 2 Miyanohigashi, Koshoin, Minami-ku, Kyoto 601-8510, Japan
(Received 3 February 2011; Accepted 14 August 2011; Published 17 September 2011)

Internal-detector electron holography is one of the atomic resolution holography methods to analyze local structures around specific elements by reconstructing 3D atomic images, and it can measure the holograms with table-top electron microscopes. Since the internal-detector electron holography uses electron beams as holographic waves, understanding of behaviors of the electron beams in solids is important to optimize the performance of the internal-detector electron holography. Here, we defined an evaluation function for the hologram measurements, $f(E)$, which is obtained from characteristic X-ray intensity and the holographic amplitude with Monte Carlo simulation. Using this formula, the best electron energy for the Ti-K hologram from SrTiO$_3$ bulk was estimated to be 0.5 keV above the Ti-K X-ray ionization energy. Next, the best energies of the electron beam for Pt thin film was calculated with varying its thickness since the quality of holographic data are degraded by the characteristic X-rays from the deep region of the sample due to breaking of the coherence of the incident electron beams. The energy profile of $f(E)$ for the thin film is quite different from that for bulk. The estimated energies were as wide as 5.0-10.0 keV above the Pt-M X-ray ionization energy. Moreover, the best thickness of thin film for hologram measurement was obtained to be about 10 nm, which is close to the inelastic mean free path of the electron in the sample.

[DOI: 10.1380/ejssnt.2011.334]

Keywords: Scanning electron microscopy (SEM); X-ray emission; Photoelectron diffraction; Surface structure, morphology, roughness, and topography; Nano-scale imaging, measurement, and manipulation technology

I. INTRODUCTION

In order to investigate the local structure analysis around the specific elements, X-ray absorption fine structure (XAFS) [1] or extended electron energy loss fine structure (EELFS) [2] method has been widely used. But from these methods, only radial distribution function up to 2nd or 3rd neighboring atoms can be obtained. Therefore, atomic resolution holography [3], which makes it possible to analyze the local structure around the specific elements within 2 nm by reconstructing 3D atomic image has attracted many attentions of researchers.

Holography which records phases and intensities of the scattered waves from an object was proposed by Gabor to improve the resolution of the electron microscope [4]. Szöke pointed out that interference patterns of the photoelectrons or fluorescent X-rays outside samples were same as atomic resolution holograms [5]. Until now, many reports on the atomic resolution holography have been published. For example, photoelectron holography [6–9] can analyze the structure of local environments of adsorbates [7]. X-ray fluorescence holography [10–14] can investigate quasicrystal structure [11] or a phase transition of a shape memory alloy [13].

However, their applications have been limited since these conventional atomic resolution holography methods
the sample. The scattered and non-scattered photoelec-
gular distribution of the photoelectron intensities around
with the non-scattered photoelectron wave, forming an-
is scattered by neighboring atoms and then it interferes
from a target atom. A part of the photoelectron wave
irradiated onto a sample and the photoelectrons excited
toelectron holography as shown in Fig. 1(a), X-rays are
detector electron holography, respectively. In the pho-
tventional photoelectron holography and (b) the internal-
for the hologram measurements,
need synchrotron radiation facilities. In order to overcome
this problem, we proposed the internal-detector electron holography [15–17], which is a time-reversed version of
the conventional photoelectron holography. Moreover, we
demonstrated the internal-detector electron holography
using a SEM with a SrTiO
In the experiment of the internal-detector electron holography, irradiated electrons penetrate into the sample
with scattering and excite the characteristic X-rays from
an element. The angular dependence of the incident electron beam on the characteristic X-ray intensity becomes
a hologram with atomic resolution. Therefore, we have
to understand the electron behavior inside the sample to
optimize the performance of the internal-detector electron holography. Here, we defined an evaluation function
for the hologram measurements, \( f(E) \), and estimated the
best energy of the incident electron for bulk and thin film
samples with Monte Carlo simulation. Moreover, since
the amplitude of the holographic signal closely relates
with the thin film thickness, we also estimated the best
thickness of the sample.

II. THEORY

Figure 1 shows the principle charts of (a) the con-
ventional photoelectron holography and (b) the internal-
detector electron holography, respectively. In the pho-
toelectron holography as shown in Fig. 1(a), X-rays are
irradiated onto a sample and the photoelectrons excited
from a target atom. A part of the photoelectron wave
is scattered by neighboring atoms and then it interferes
with the non-scattered photoelectron wave, forming an-
gular distribution of the photoelectron intensities around
the sample. The scattered and non-scattered photoelec-
trons serve as the object and reference waves in holo-
graphy, respectively, and the interference pattern becomes
a hologram, which has information of the 3D atomic ar-
angement around the target atom. On the other hand, as
shown in Fig. 1(b), the internal-detector electron hologra-
phy which is the time-reversed version of the conventional
photoelectron holography, uses the interference of the in-
cident electron beam. When an electron is irradiated to
the solid, a part of the electron wave is scattered by atoms
near the target atom and then it interferes with the non-
scattered electron wave around the target atom. In this
case, the scattered and non-scattered electrons also serve
as the object and reference waves in holography, respec-
tively. Since the amplitude of the electron interference
pattern changes by the direction of the incident electron
beam, the angular dependence of the characteristic X-ray
intensity from the target atom becomes a hologram, which
is equivalent to the conventional photoelectron hologram.

In the internal-detector electron holography, the elec-
tron beams must be irradiated to the sample with en-
ergy higher than the X-rays ionization energy of the tar-
get atom. The electrons penetrate into the deep region
with multiple scattering and generating the characteristic
X-rays. Figure 2 shows a schematic illustration of elec-
tron behavior in a solid. As mentioned in Fig. 1(b), the internal-detector electron holography uses elastically scat-
tered electrons (object wave), which are coherent with the
incident electrons and interfered with the incident elec-
trons (reference wave). The characteristic X-rays, whose
intensity is modulated by the interference between scat-
tered and non-scattered electron waves, contribute to the
hologram. While, some electrons are inelastically scat-
tered, and lose their energies and coherences. If such
electrons excited the characteristic X-rays, they become
the background, which do not contribute to the hologram.
Therefore, total X-ray intensity \( I_t \) can be divided into \( I_h \)
and \( I_{bh} \) which contributes to the hologram and the back-
ground, respectively.

In principle of the electron scattering, \( I_h(t) \) at depth \( t \)
can be written by,

\[
I_h(t) = I_{h0} \times \exp \left( -\frac{t \times \cos \theta}{l} \right),
\]

where \( \theta \) is the incident angle of the electrons and \( l \) is the
inelastic mean free path. Spence and Koch mentioned
that electron energy slightly higher than the X-ray ioniza-
!
Q is written by energies by multiple inelastic scatterings. Since the low than the ionization energy even though they lose their of the inelastically scattered electrons with higher energy comparable to or more than the $I$ loss of their energy [16]. Therefore, under this condition, electrons hardly excite the characteristic X-rays due to the detection angle of the electrons trajectories.

FIG. 4: (a) Simulation of 10,000 electron trajectories in SrTiO$_3$ sample at 6.0 keV with CASINO. Blue pattern shows the electrons which penetrate into the solid with decreasing their energies until less than 50eV and red pattern shows the backscattered electrons. (b) Ti-K characteristic X-ray intensity curve of a SrTiO$_3$ sample simulated at 6.0 keV, 10,000 electrons trajectories.

Here, total ionization cross section of K-shell ionization $Q$ is written by

$$Q = \frac{2\pi e^4}{V_K E} \times 0.35 \times \ln \frac{4E}{1.65 + 2.35 \times \exp \left( 1 - \frac{E}{E_t} \right)} V_K,$$

(2)

where $E$ is the kinetic energy of the incident electron and $V_K$ is the ionization energy of the K shell, respectively [19]. In Fig. 3, the curve shows the values of $Q$ calculated for Ti-K shell ionization. Since the cross section of the inner-shell ionization at the electron energy just above the threshold is very small, the statistical error of $I_t$ and $I_b$ is very large. In order to improve the statistical error of X-ray intensity, the electron energy should be much higher than threshold. Under this condition, total characteristic X-rays are generated strongly owing to the large total ionization cross section. However, $I_{th}$ is comparable to or more than the $I_b$ since there are a lot of the inelastically scattered electrons with higher energy than the ionization energy even though they lose their energies by multiple inelastic scatterings. Since the low ionization energy is efficient for the measurement of the internal-detector electron holography since inelastically scattered electrons hardly excite the characteristic X-rays due to the loss of their energy [16]. Therefore, under this condition, $I_t$ is approximately equal to $I_b$.

$\frac{I_b}{I_t}$ means the low holographic amplitude, the hologram measurement under this condition is difficult. Therefore, we have to determine the electron energy to optimize the performance of the internal-detector electron holography by understanding the electron behaviors in solids.

III. SIMULATION

A. In the case of SrTiO$_3$

To simulate the behavior of the electron beams in the sample, using the Monte Carlo simulation program of CASINO [20, 21], we calculated the characteristic X-ray yields with changing electron energies and estimated the best energy of the incident electron beams. Figure 4(a) shows an example of electron trajectories in a bulk SrTiO$_3$ when 6.00 keV electron beams are irradiated at the incident angle of the electrons $\theta = 0^\circ$. In Fig. 4(a), 10,000 electrons penetrate into the SrTiO$_3$ until their energies decrease to 0.05 keV (in blue) or are backscattered from the surface of the sample (in red). Figure 4(b) shows the depth dependence of the intensity of the Ti-K characteristic X-rays, the solid and dashed curves indicate $I_t$ and $I_b$ respectively. The $I_t(t)$ indicates the intensity of characteristic X-rays generated at depth $t$, which was calculated by CASINO and $I_b(t)$ was calculated by Eq. (1).

As mentioned above, for the hologram measurement, $I_b/I_t = \Sigma I_b(t)/\Sigma I_t(t)$ means the holographic amplitude, and must be larger than the statistical error $\sqrt{N}/N = 1/\sqrt{N}$, where $N$ is total counts of the detected characteristic X-rays, and here, $N = I_t$. If we define a function, $f(E)$:

$$f(E) = \frac{I_b/I_t}{1/\sqrt{I_t}} = \frac{I_b}{\sqrt{I_t}},$$

(3)

the hologram measurement becomes easier with increasing $f(E)$.

Using a bulk sample, the results of $f(E)$ do not vary with the changes of the incident angle $\theta$, therefore we discuss about $f(E)$ only in the case of $\theta = 0^\circ$. Figure 5
Fig. 5: The solid and dashed curves show the energy dependence of $I_t$ (right axis), and $I_h$ (left axis) respectively. (a): 5.0-10.0 keV. (b): 5.0-5.2 keV.

displays the calculated $f(E)$ for the Ti-K hologram from SrTiO$_3$ in the region of 5.0-15.0 keV. In Fig. 5, the maximum of $f(E)$ appears at around 5.5 keV, which is around 0.5 keV above the Ti-K X-ray absorption edge of 4.96 keV. In Fig. 6, the solid and dashed curves show the energy dependences of $I_t$ and $I_h$ respectively. The increments of $I_t$ is larger than that of $I_h$ as a whole. Moreover, $I_h$ increases rapidly in the low energy, then increases slowly in the higher energy regions, however $I_t$ is vice versa. Consequently, $f(E)$ has the maximum at around 5.5 keV, which is the best energy for hologram measurement. The value of $I_h/I_t$ is 0.326 at 5.5 keV. The holographic amplitude in the X-ray fluorescence holography is about 0.1% of the background, and the holographic amplitude in the photodetector holography is about 15%. Therefore, the hologram of the internal-detector electron holography is easy to observe like the conventional photodetector holography. Actually, in Ref. [18] the holographic amplitude is approximately 12% to their background intensities at 6.00 keV.

We also calculated $f(E)$ of the Sr-K hologram in order to compare that of the Ti-K hologram since the electron behavior depends on their kinetic energies. Figure 7 displays the calculated $f(E)$ of the Sr-K hologram from SrTiO$_3$ in the region of 16.0-24 keV. In Fig. 7, the maximum of $f(E)$ appears at around 16.4 keV, which is 0.30 keV above the Sr-K X-ray absorption edge of 16.1 keV. $I_h/I_t$ is 0.0703 at 16.4 keV, and this value is smaller than that of Ti-K, because of the long inelastic mean free path (19.52 nm [22]) at 16.4 keV.

B. In the case of Pt thin film

In the case of a bulk sample, characteristic X-rays generated from the deep region behave as backgrounds owing to the inelastically scattered electrons. On the other hand, in the case of thin film, the background should be suppressed since the inelastically scattered electrons penetrate into the substrate and the characteristic X-rays generated in the substrate do not influence the hologram. In this section, to know how effectively the hologram can be measured for a thin film sample, we calculated $f(E)$ for different thicknesses.

Figure 8 displays the calculated $f(E)$ of the Pt-M hologram from 0.1-100 nm thick Pt thin films in the energy region of 2.0-15.0 keV at $\theta = 0^\circ$. As shown in Fig. 8(a), $f(E)$ of 0.1 nm Pt film is approximately 0.14 of that of 5 nm Pt film and $f(E)$ of 1 nm Pt film is approximately 0.5 of that of 5 nm Pt film as a whole. Since $I_h$ is proportional to the film thickness and the characteristic X-rays which do not contribute to the hologram are hardly generated, $f(E)$ is proportional only to the root of the thickness. For 5 nm Pt film, the maximum of $f(E)$ appears at 10.5 keV. In Figs. 8(a) and (b), $f(E)$ of 10 nm Pt film is larger than that of 5 nm Pt film above 9.0 keV, and is larger than that of 20 or 50 nm Pt film as a whole. Moreover, the profile of $f(E)$ for 100 nm Pt film is similar to that for a bulk sample as shown in Fig. 5. These results indicate that the good thickness value of the hologram measurement is 10 nm.

However, for a thin film sample, the value of $f(E)$ depends on $\theta$ unlike a bulk sample since at the high $\theta$ the path distance electron beams traveling in the Pt film become long and the background X-rays by the inelastically scattered electrons increase. As a result, in the high $\theta$, the behavior of the incident electrons is similar to that in the bulk. Therefore, we calculated $f(E)$ of the Pt-M holo-
gram from 1-100 nm Pt films in the region of 2.0-15.0 keV at $\theta = 0, 15, 30, 45, 60, 75^\circ$. Figure 9 shows the calculated $f(E)$ for 10 nm Pt film at $\theta = 0-75^\circ$. As shown in Fig. 9, the value of $f(E)$ decreases as a whole with increasing $\theta$ since $I_{nh}$ and $I_t$ increases with increasing $\theta$. Indeed, the profiles of $f(E)$ at $\theta = 60, 75^\circ$ are similar to that of a bulk sample as shown in Fig. 5. In order to compare the value of the calculated $f(E)$ for 1-100 nm Pt films simply, the values of $f(E)$ at $\theta = 0-75^\circ$ for each thickness are averaged. Figure 10 shows the averaged $f(E)$ for 1-100 nm Pt films. In Fig. 10, $f(E)$ of 10 nm Pt film is also larger than the others in a whole the energy region. Therefore, the adequate thickness of Pt film is estimated to be around 10 nm and this result is similar to that obtained from Fig. 8 although, $f(E)$ in Fig. 10 is smaller than that in Fig. 8. Here, the inelastic mean free path $l$ in Pt is 9.25 nm at 10.0 keV [22]. If the film was thicker than , more characteristic X-rays excited by inelastically scattered electrons emit from the region deeper than $l$ and this decreases the value of $f(E)$. Therefore, the best thickness of the sample is close to the inelastic mean free path in the Pt thin film. However, the maximum of $f(E)$ for a thin film sample does not appear clearly unlike a bulk sample since $I_{nh}$ of thin film does not increase largely at the high energy.

**IV. CONCLUSION**

In this paper, we estimated the best energy of the incident electron for the internal-detector electron holography by the Monte Carlo simulation program, CASINO. We defined the evaluation function for the hologram measurement and estimated the best energy of the incident electron beams for bulk and thin film samples. In the case of a SrTiO$_3$ bulk sample, the best energy of the incident electron is several hundred eV higher than the ionization energy of the target atom. On the other hand, in the case of a thin film sample, the background is suppressed since the multiple inelastically scattered electrons transmitted into the substrate and the characteristic X-rays generated in the substrate do not influence the hologram. Therefore, there is not the obvious peak of $f(E)$ for ultra thin films. Moreover, the best thickness was estimated to be close to the inelastic mean free path of the electron. In the future, we have to measure the holograms of Pt thin film with various thicknesses evaporated on MgO at various energies in order to confirm these results. Moreover, by measuring the applications such as the ferromagnetic semiconductor, the wide possibility of the internal-detector electron...
holography must be demonstrated.

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