Precise Evaluation of Charging Effect on SiO₂ Particles by Simulation of Holograms *

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The charging effect due to the electron irradiation in spherical SiO₂ particles of 250 nm in diameter has been analyzed by electron holography. Electron holograms of a charged SiO₂ particle on the side surface of the carbon film were simulated taking into account the electric shielding effect due to the carbon film. In order to compare the observed hologram and the simulated holograms quantitatively, the residual index between the observed and simulated images was evaluated. Through the quantitative analysis taking into account the shielding effect with the limited side surface area of the carbon film, the amount of the charge was evaluated to be $3.38 \times 10^{-17}$ C for the current density of incident electron beam at 0.24 A/m² without an objective aperture.

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1. Introduction

Since transmission electron microscopes with a field emission gun have been developed, electron holography technique¹,² is now expected to apply for characterizing a variety of materials. Electron holography is a unique technique especially for observing electromagnetic fields in electronic and magnetic materials. The electromagnetic fields can be recorded by the phase shift against periodic interference fringes on an electron hologram. The intensity of the electron hologram $I$ is given by

$$I = 1 + A² + 2A \cos \left( \phi + 2\pi \frac{\alpha}{\lambda} \right)$$  \hspace{1cm} (1)

where $A$ and $\phi$ are real functions which describe the amplitude change and the phase shift due to the electromagnetic fields, respectively. λ is the wavelength of an electron and $\alpha$ is a tilted angle of electron wave due to the voltage of a biprism. The phase shift is obtained from the electron hologram directly or extracted by using the optical process or the Fourier transform. In the reconstruction process of the latter case, the electromagnetic fields can be imaged by evaluating equiphaseline which are equipotential lines of the projected potential.⁴⁻⁷

On the other hand, it is well known that insulate specimens are electrically charged under the strong electron irradiation, and this charging effect can be also evaluated by electron holography. Actually the charging effect in latex⁸⁻¹⁰ and SiO₂¹⁰ particles has been discussed through electron holography experiments. In these reports, two experimental conditions were utilized as shown in Figs. 1(a) and (b). In the case of Fig. 1(a), the latex particle is on the carbon film and the electron beam was incident perpendicular to the carbon film. In the other case, the latex particle or the SiO₂ particle is on the side surface of a copper grid or carbon film as shown in Fig. 1(b). In the latter case, the area of the side surface in the carbon film is assumed to be wide enough to apply the charge image method.⁵ In both cases, the estimation of the amount of the charge due to the electron irradiation had been reported."⁻¹⁰

In this paper, by electron holography, we investigate the charging effect in a spherical SiO₂ particle of 250 nm in diameter on the side surface of a thin carbon film, which corresponds to the solid line in Fig. 1(b). Differing from the previous paper¹⁰ where the amount of the charge was estimated from the phase shifts observed at a few points of the electron hologram, the image simulation of electron holograms is carried out taking into account the charging effect. From the observed hologram and the simulated holograms, the residual index is evaluated as a function of the electric shielding effect and the amount of the charge. By minimizing the residual index, the amount of the charge is accurately determined.

2. Experimental Procedures

Amorphous SiO₂ particles have a spherical shape and their average diameter is about 250 nm. The particles mixed with butyl alcohol were dropped on a carbon microgrid for TEM observation. Since electron holograms can be obtained by interfering the wave passing through the object and the vacuum,
the fairly large vacuum region is necessary near the object in the microgrid. Thus, the carbon microgrid was scratched for making a hole before dropping the alcohol mixed with particles.

Electron holography on the electrical charging effect was carried out with a JEM-3000F TEM with an accelerating voltage of 300 kV. The microscope is equipped with a thermal field-emission gun and a biprism. The biprism and two earth potential electrodes are positioned between objective lens and intermediate lens. In this study, the magnification of image was set to be 4000 times and the voltage of the biprism was set to be 45 V. Electron holograms recorded on conventional EM films were digitalized using a Nikon-LS-3510AF film scanner to be 45 V. Electron holograms recorded on conventional EM films were digitalized using a Nikon-LS-3510AF film scanner with resolution of 8 µm/pixel. Reconstructed phase images were obtained from the digitized holograms with the Fourier transform. The phase shift \( \phi(x, y) \) due to a SiO\(_2\) particle is represented by \( \cos(2\pi \phi(x, y)) \) in the reconstructed phase images.

3. Results and Discussions

Figure 2(a) shows an electron hologram of a charged SiO\(_2\) particle of 250 nm in diameter observed at 300 kV. Due to the charging effect of the particle, interference fringes curve clearly near the surface of the SiO\(_2\) particle as indicated by arrows. Figure 2(b) shows a digital diffractogram obtained from the hologram of Fig. 2(a) with the Fourier transform. Two sidebands at the upper part and lower part are seen in Fig. 2(b). Figure 2(c) shows a reconstructed phase image obtained with the area circled in Fig. 2(b) by the inverse Fourier transform. While the reconstructed phase image of Fig. 2(c) is shown with no phase amplification, the phase of the reconstructed phase image in Fig. 2(d) is amplified by 4 times. In Fig. 2(d), the electric field due to the charging effect around the particle is clearly seen. The reconstructed phase images indicate the phase shift due to not only the inner potential inside the particle but also the electric field around the particle. It is noticed that the electric field around the particle is not symmetric against the center of the particle. The phenomenon is considered to result from the electric shielding effect due to the supporting carbon film.\(^9\) Here, we quantitatively evaluate the electric shielding effect and the amount of the charge on the SiO\(_2\) particle. Although the reconstructed phase image can be used for evaluation of the charging effect, it is sensitively affected by the area of the digital diffractogram to be selected and the accuracy of the Fourier transform. Thus, we analyze the electron hologram with image simulation to determine the amount of the charge in this paper. In the previous paper,\(^6\) the phase shift due to the electric field is given by

\[
\phi = \frac{\sigma q}{4\pi \varepsilon_0} \int_0^\infty \left( \frac{1}{\sqrt{(x-a)^2+y^2+z^2}} - \frac{1}{\sqrt{(x+a)^2+y^2+z^2}} \right) dz
\]

\[
\sigma = \frac{2\pi}{\lambda E(1+\sqrt{1-\beta^2})}, \quad \beta = v/c
\]

where \(\sigma\) is an interaction constant, which depends on the accelerating voltage of an electron microscope. \(E\) and \(\lambda\) are the accelerating voltage and the electron wavelength, while \(v\) is the electron velocity and \(c\) is the light velocity. In this paper, differing from the previous assumption,\(^7,9\) the partial shielding effect is assumed, and thus the equation for the phase shift contains partial shielding factor \(C\) as given by

\[
\phi = \frac{\sigma q C}{4\pi \varepsilon_0} \int_0^\infty \left( \frac{1}{\sqrt{(x-a)^2+y^2+z^2}} - \frac{1}{\sqrt{(x+a)^2+y^2+z^2}} \right) dz
\]

\[
+ \frac{\sigma q (1-C)}{4\pi \varepsilon_0} \int_0^\infty \frac{1}{\sqrt{(x-a)^2+y^2+z^2}} dz
\]

\[
= \frac{\sigma q}{4\pi \varepsilon_0} \int_0^\infty \left( \frac{1}{\sqrt{(x-a)^2+y^2+z^2}} - \frac{C}{\sqrt{(x+a)^2+y^2+z^2}} \right) dz
\]

where \(\varepsilon_0\) is the dielectric constant of vacuum and \(q\) is the quantity of the electric charge on the SiO\(_2\) particle. In the above equation, for \(C = 1\), the charge is assumed to be shielded perfectly due to the carbon film and the equation is the same as eq. (2). On the other hand, for \(C < 1\), the charge is assumed not to be shielded perfectly. Taking into account a limited thickness of carbon film shown in Fig. 1(b), the phase shifts were calculated by varying the shielding factor \(C(\leq 1)\).

Figure 3 shows the comparison between the observed hologram and the simulated holograms. An electron hologram of Fig. 3(a) shows the enlarged hologram of a part in Fig. 2(a). Figure 3(b) shows a simulated hologram obtained only tak-
Fig. 3 (a) Observed hologram of a charged SiO$_2$ particle. (b) Simulated hologram obtained taking into account only the inner potential of a SiO$_2$ particle. (c) Simulated hologram obtained taking into account the inner potential and the charging effect. The arrows indicate the directions of interference fringes near the SiO$_2$ particle.

It is noted that the interference fringes are straight in all parts except for the inside of the particle. A simulated hologram obtained taking into account not only the inner potential but also the charging effect on the particle is shown in Fig. 3(c). Note that the interference fringes in the simulated hologram of Fig. 3(c) curve near the surface well corresponding to the observed hologram of Fig. 3(a). In order to estimate the charging effect quantitatively, we evaluate the residual index $R_{Holo}$

\[
R_{Holo} = \frac{\sum |I_{obs} - I_{cal}|}{\sum I_{obs}} \tag{4}
\]

in the square region shown in Figs. 3(a), (b) and (c). Although in the vacuum region, intensity of the observed hologram can be directly compared with the normalized intensity of the simulated hologram, intensity of the observed hologram inside the particle gradually changes due to the absorption effect and it is difficult to compare the observed hologram with the simulated holograms quantitatively. Thus, we evaluated the residual index only in the vacuum region in this paper. The residual index obtained from the holograms in Figs. 3(a) and (b) is 0.33. Figures 4(a) and (b) show the residual index obtained from the observed hologram and the simulated holograms as a function of the amount of the charge and the shielding factor, respectively. The amount of charge and the shielding factor were determined to minimize the residual index. Here we get the minimum value of the residual index 0.061 in these calculations. And, it is concluded that the amount of the charge due to the electron irradiation was determined to be $3.38 \times 10^{-17}$ C for the current density of incident electron beam at 0.24 A/m$^2$ without an objective aperture. Also, the shielding factor is determined to be 69.9%.

4. Conclusions

Quantitative analysis of the charging effect on SiO$_2$ particles of 250 nm in diameter on the side surface of the carbon film with electron holograms is summarized as follows.

(1) In order to compare the observed hologram with the simulated holograms quantitatively, the residual index is successfully introduced and evaluated to determine the charging effect on a SiO$_2$ particle.

(2) The amount of the charge on the SiO$_2$ particle due to the 300 keV electron irradiation of 0.24 A/m$^2$ is determined to be $3.38 \times 10^{-17}$ C under the experimental condition without an objective aperture.

(3) Through the quantitative analysis, it is found that the electric shielding effect of the carbon film is quantitatively evaluated for the charged spherical SiO$_2$ particle, and the shielding factor is determined to be 69.9%.

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