https://doi.org/10.2352/EI.2022.34.14.COIMG-248

Accurate measurement of charge density in nanoscale particles using an aperture optimization of Fourier based phase reconstruction

Takuma Okada, Yoshihiro Midoh, Koji Nakamae, Noriyuki Miura; Osaka University; Suita, Osaka, Japan

Abstract

Electron holography is used to observe the nanometer-scale electromagnetic field distributions of electronic and magnetic materials. The signal-to-noise ratio of electron hologram decreases when the electron beam irradiation is reduced to avoid unnecessary charging and damage. Noise in the hologram causes phase errors. To obtain accurate phase information, we propose an aperture optimization of Fourier based phase reconstruction. Our method effectively separates the signal from the noise using an extended Fourier ring correlation. From the experimental results using a simulated electron hologram with low signal-to-noise, it was found that the proposed method reduces the phase error to 41% of the conventional method. When applied to a real hologram, the proposed method achieved smoother phase reproduction.

Introduction

Electron holography, which was invented by Gabor [1] and observed for the first time by Haine and Mulvey [2], is used as a method to observe the distribution of electromagnetic fields in nano-scale structures using TEM (transmission electron microscope) as shown in Figure 1. A coherent electron wave emitted from the electron source propagates to a sample. Half of the wave passes through the sample (object wave), and the other half passes through a vacuum area (reference wave). The object and reference waves are superimposed by using a biprism filament under the object plane and then an interference fringe pattern is formed. The object wave is accelerated or decelerated depending on the internal structure and electric charge of the sample, and these sample conditions appear as shifts or distortions in the interference fringes of the hologram. Therefore, the electric field distribution is visualized by detecting the phase shift of the interference fringe pattern in the hologram.

To measure a minute phase change accurately, it is desirable to improve the image quality of the hologram, i.e., the contrast and signal-to-noise ratio (SNR) of the interference fringes [3]. To obtain a hologram with a good SNR, it is possible to increase the beam current and extend the exposure time. These approaches may, however, be difficult in terms of sample damage, contamination, and charging. It is expected to improve the image quality of holograms by image processing techniques. Conventional noise reduction methods such as Gaussian filtering and non-local means filter are widely used [4]. These methods assume that the noise component is relatively high frequency compared to the true signal component. On the other hand, electron holograms have relatively high frequency component to measure the electromagnetic field distribution of the sample with the highest resolution possible. Therefore, there is a risk of losing or incorrectly extracting the sample information by the noise reduction. In recent years, noise reduction methods using deep learning have been proposed [5]. However, for electron holograms that may contain unknown physical phenomena, the challenge is to acquire clean teacher data for training and to guarantee the reliability of black-box predictions.

In this paper, we propose a new method to incorporate noise reduction into Fourier transform-based phase reconstruction, which is mainly used for phase extraction from electron holograms. Figure 2 shows a procedure of the Fourier transform-based phase reconstruction for electron holograms [6]. First, a fast Fourier

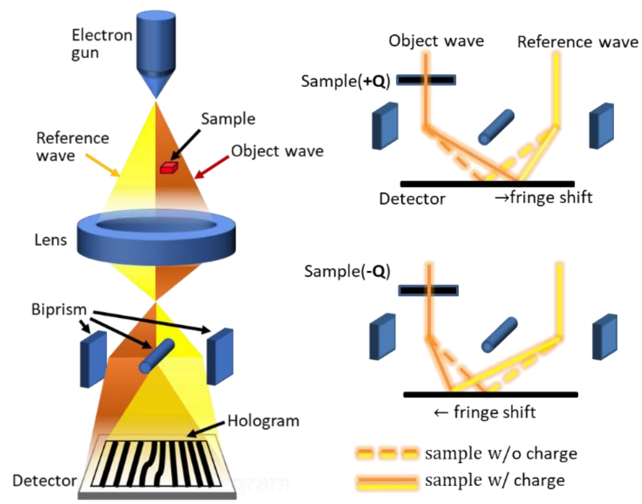

Figure 1. A schematic illustration of electron holography. The emitted electron wave produces an interference fringe pattern depending on the state of the sample.

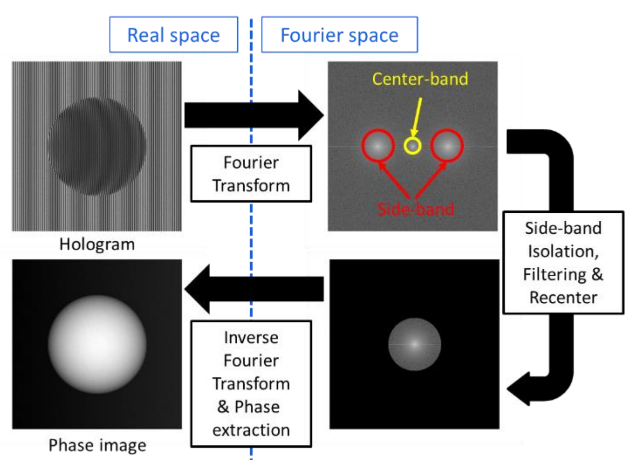

Figure 2. A procedure for generating a phase image using the Fourier transformed-based phase reconstruction for an electron hologram.

transform (FFT) of the hologram is performed. The resulting complex image consists of the autocorrelation (center band) and two mutually conjugated sidebands. Since sample information to be extracted is contained in the sideband, only one sideband is selected by applying a low-pass filter (ideal low-pass filter, Butterworth filter etc.) centered on the chosen sideband. The shape of the low-pass filter is called an aperture. Next, the selected sideband is re-positioned to the center of the complex image and the inverse FFT is applied. The resulting image is complex-valued and thus the phase distributions are obtained by an arc tangent operation and phase unwrapping to connect phase gaps (2π phase jump) smoothly.

The aperture is generally isotropic, and its radius is one-third the distance between the center band and the sideband peaks [3]. In fact, there is a trade-off between the aperture size and the image quality of the phase image. The larger the aperture size, the higher the resolution of the phase image, but the lower the SNR. For holograms with low SNR, larger apertures will generate more phase error to pick up noise around the isolated sideband. However, the high frequency components of the signal must also be contained within the aperture to extract the phase as accurately as possible.

Therefore, we try to design an optimal anisotropic aperture for the input hologram to remove noise in Fourier space while retaining the valid signal about the sample. Our proposed method separates the signal and noise around the sideband in Fourier space by extending Fourier ring correlation (FRC), which is used to evaluate the resolution of electron tomograms. In addition, automatic region of interest (ROI) extraction is performed to minimize artifacts caused by periodic disturbance of the edge of a hologram during Fourier transform as shown in Figure 3.

This paper gives an overview of the proposed method and shows comparison results with conventional methods. The outline of this work is organized as follows. Methods introduce the automatic ROI extraction as the preprocessing and the detailed procedure for the aperture optimization of Fourier based phase reconstruction. Results are devoted to simulation and experimental results. Finally, the conclusion is drawn in Concluding Remarks.

Methods

Automatic ROI extraction

Electron holograms are generally observed in such a way that the interference fringes are oblique to balance the horizontal and vertical resolution of the resulting phase image. Therefore, due to the discontinuity of the edges, a wide band of frequency components in the direction along horizontal, vertical, and the fringe angle θ may be generated as artifacts during the Fourier transform as shown in Figure 3. To reduce artifacts and avoid calculating unnecessary regions, the automatic ROI extraction is performed as the preprocessing. Figure 4 shows the procedure. First, the fringe angle is obtained from the angle between the origin and sideband peaks in Fourier domain. Next the width W of the interference fringes is determined by finding edges of the background and the fringe pattern from the average line profile in the vertical direction of the rotate hologram. Finally, a region of size W x W is extracted as ROI from the rotated hologram.

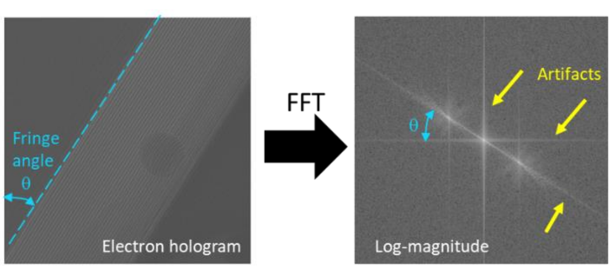

Figure 3. Artifacts in Fourier domain caused by periodic disturbance of the edge of a hologram during Fourier transform.

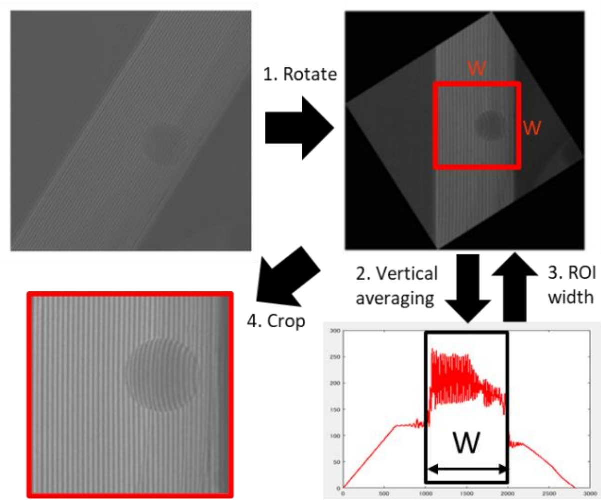

Figure 4. The procedure of the automatic ROI extraction.

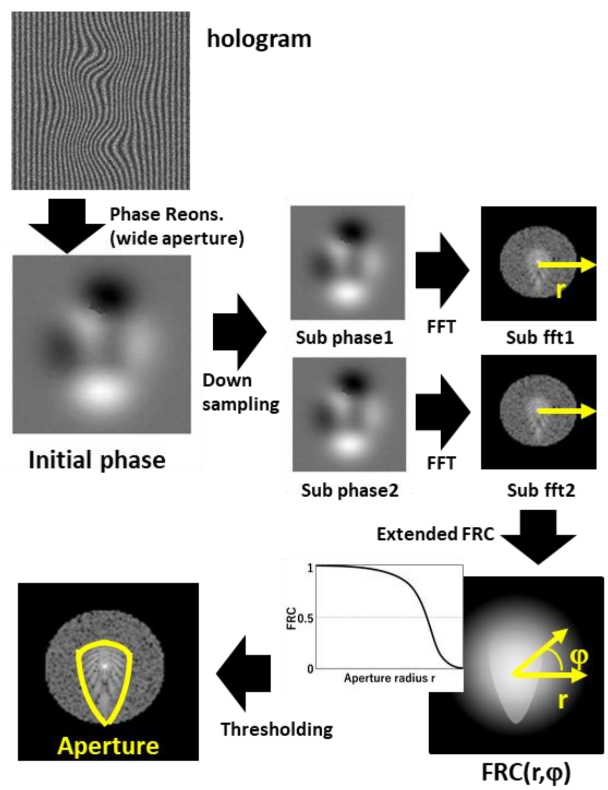

Figure 5. The procedure of the aperture optimization for the Fourier based phase reconstruction.

Aperture optimization

We design a suitable aperture to be used instead of a low-pass filter in Fourier based phase reconstruction. To separate the signal from the noise in the Fourier domain, we use Fourier ring correlation (FRC). The FRC measures the normalized cross-correlation coefficient between two images at different spatial frequencies. FRC is given by the following equation,

$$\mathrm{FRC}(\mathrm{r}) = \frac{\sum_{\mathrm{r}} \mathrm{F}_1(\mathrm{r}) * \mathrm{F}_2(\mathrm{r})^*}{\sqrt{\sum_{\mathrm{r}} |\mathrm{F}_1(\mathrm{r})|^2 \sum_{\mathrm{r}} |\mathrm{F}_2(\mathrm{r})|^2}} \tag{1}$$

where r is radius of the polar coordinate system, $F_1(r)$ and $F_2(r)$ are complex images obtained by applying the Fourier transform to two images, and * denotes complex conjugate. When the two images have an identical signal corrupted by white random noise, the FRC is close to 1 in the low frequency region containing the sample information and approaches 0 at higher frequencies because uncorrelated noise is dominant.

We propose an extended FRC that considers the argument φ of the polar coordinate system as follows.

$$\mathrm{FRC}(\mathrm{r}, \varphi) = \frac{\sum_{\mathrm{r},\varphi} \mathrm{F}_1(\mathrm{r},\varphi) * \mathrm{F}_2(\mathrm{r},\varphi)^*}{\sqrt{\sum_{\mathrm{r},\varphi} |\mathrm{F}_1(\mathrm{r},\varphi)|^2 \sum_{\mathrm{r},\varphi} |\mathrm{F}_2(\mathrm{r},\varphi)|^2}} \tag{2}$$

where plus or minus a few degrees of the argument of interest are considered in the summation operation.

Figure 5 shows a procedure of the proposed method. First, the given hologram is filtered with a wide low-pass filter and then its phase image is obtained using the conventional Fourier based reconstruction. Next, we generate two half-size phase images by sampling the columns and rows with even and odd numbers, respectively. These two images are assumed to have approximate identical signal and independent noise. The FRC map is obtained by calculating FRC in both different radii and arguments. The spatial frequency and orientation with small FRC values are more likely to be noise. Therefore, we extract the region where the FRC value is greater than 0.5 as a closed curve. Using the extracted anisotropic aperture, the phase reconstruction is again applied to the input hologram.

Results

Simulation results

We evaluated the validity of the proposed method using a simulated hologram of an uncharged SiO2 particle as shown in Figure 6 [7]. The noise in the electron hologram can be considered as Poisson distribution due to single electron events. We applied four phase reconstruction methods to a noisy hologram with Poisson noise. Figures 7 shows (a) ground-truth phase image and reconstructed phase image by each method; (b) low-pass filter for the raw hologram (conventional method), (c) anisotropic aperture for the raw hologram, (d) low-pass filter for the extracted ROI hologram, and (e) anisotropic aperture for the extracted ROI hologram (our method), respectively. Here, application results to the raw hologram are phase images obtained by applying it to the entire simulation hologram and then cropping to be in the same region as the ROI. It was confirmed that the variation in the phase image was reduced and a smoother phase was obtained by using the anisotropic aperture obtained by the proposed method in comparison with the conventional low-pass filter. The extraction of ROIs was also found

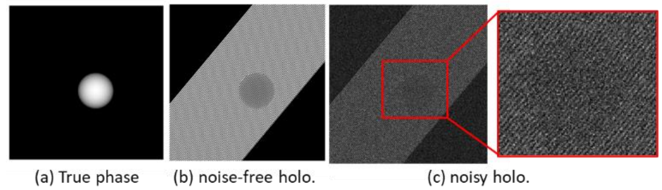

Figure 6. Simulated electron holograms for an SiO2 particle without charge; (a) ground-truth phase image, (b) a noise-free hologram, and (c) a hologram with Poisson noise and its magnified view.

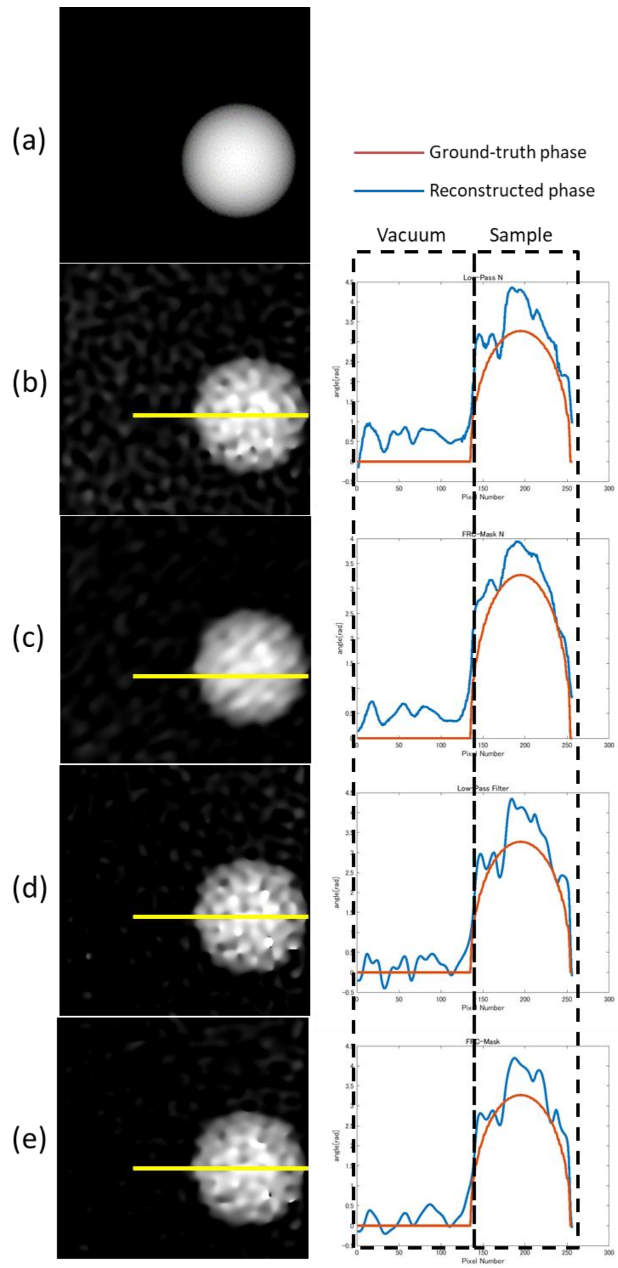

Figure 7. A comparison of reconstructed phase images and line profiles along the center of the sample; (a) ground-truth phase image, (b) low-pass filter for the raw hologram, (c) anisotropic aperture for the raw hologram, (d) low-pass filter for the extracted ROI hologram, (e) anisotropic aperture for the extracted ROI hologram.

Table 1. A comparison of the mean absolute error (MAE) between the ground-truth phases and the reconstructed phases by using each method.

Figure	Target	Filter type	MAE	Error ratio
7(b)	Raw holo.	Low-pass	0.430	100%
7(c)		Anisotropic aperture	0.241	56%
7(d)	ROI holo.	Low-pass	0.207	48%
7(e)		Anisotropic aperture	**0.177**	**41%**

to be effective in reducing the phase bias. In order to quantitatively compare each method, the mean absolute error (MAE) between the reconstructed phase $\phi(x, y)$ and the ground-truth phase $\phi_{gt}(x, y)$ was calculated by the following equation.

$$\mathrm{MAE} = \frac{1}{W^2}\sum_{i=1}^{W}\sum_{j=1}^{W}\left|\phi(i,j)\text{-}\phi_{gt}(i,j)\right| \quad (2)$$

Table 1 shows the results of calculating the MAE of the phase images obtained by each method. The proposed method achieved a 41% reduction in MAE compared to the conventional method. Since the proposed method can extract a phase closer to the ground-truth, it is expected that it is applied to low SNR holograms with the shorter electron beam exposure and the natural state of the sample without electron beam damages and charging can be observed.

Experimental results

We evaluated the validity of the proposed method using a real electron hologram of an latex particle as shown in Figure 8. The experiment was performed with Holography TEM HF-3300X. The hologram was recorded using Gatan Ultrascan camera. Figure 9 shows phase images and line profiles along the center of the sample obtained by each method, (a) low-pass filter for the raw hologram, (b) anisotropic aperture for the raw hologram, (c) low-pass filter for the extracted ROI hologram, (d) anisotropic aperture for the extracted ROI hologram (our method). In the phase images except for Figure 9(d), it can be seen that the phase unwrapping does not connect the phase gap well due to the phase noise. The proposed method was able to capture the steep phase change at the boundary between vacuum and the sample while smoothly connecting the phases.

Concluding remarks

We proposed a novel procedure of Fourier based phase reconstruction for noisy electron holograms observed at low electron dose levels to improve the limit of the phase reconstruction. Our method enables to extract ROI automatically and to design an optimal anisotropic aperture for single electron hologram with low SNR by using the extended Fourier ring correlation. We evaluated the effects of preprocessing and aperture optimization, to the simulated and real electron holograms. From simulation results, it is found that proposed method reduced the phase error to 41% compared with the conventional low-pass filter. It was also shown to be effective for the real hologram.

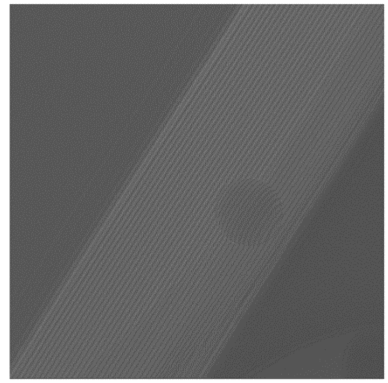

Figure 8. A real hologram of a latex particle observed by holography TEM.

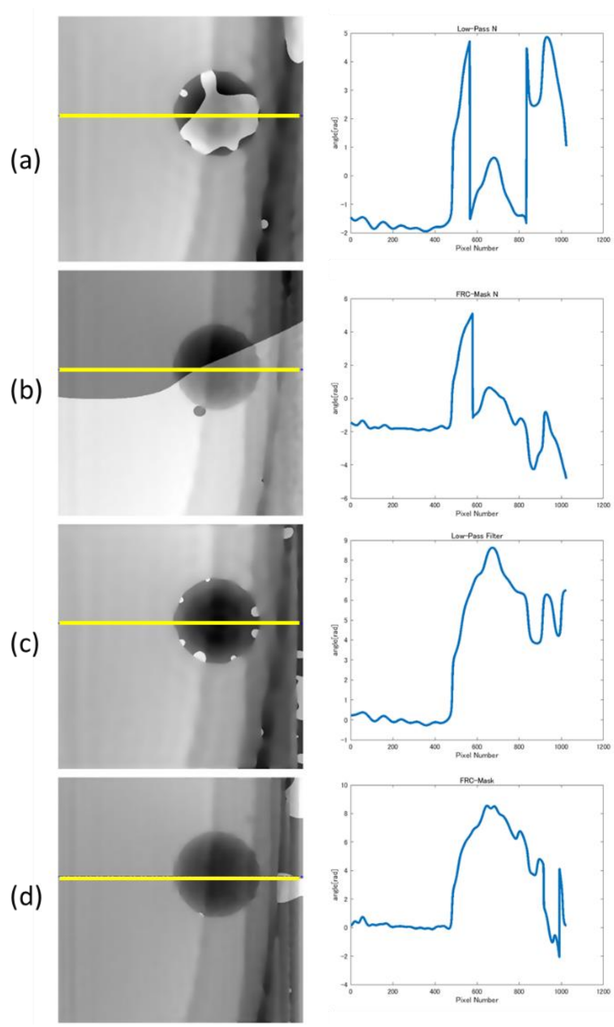

Figure 9. A comparison of phase images reconstructed by applying each method to a real hologram and line profiles along the center of the sample; (a) low-pass filter for the raw hologram, (b) anisotropic aperture for the raw hologram, (c) low-pass filter for the extracted ROI hologram, (d) anisotropic aperture for the extracted ROI hologram.

Acknowledgments

This work was supported by CREST, JST (Grant No. JPMJCR1664). We would like to gratefully and sincerely thank Prof. Yasukazu Murakami and Dr. Youngji Cho of Kyushu University for providing experimental data and their immense support.

References

[1] D. Gabor, “A new microscopic principle,” Nature, vol. 161, pp. 777–778, 1948.

[2] M. E. Haine and T. Mulvey, “The formation of the diffraction image with electrons in the Gabor diffraction microscope,” J. Opt. Soc. Am., vol. 42, no. 10, pp. 763–773, 1952.

[3] H. Lichte and M. Lehmann, “Electron holography—basics and applications,” Reports on Progress in Physics, vol. 71, no. 1, 016102, 2008.

[4] J. Roels et al., “An overview of state-of-the-art image restoration in electron microscopy," Journal of microscopy, vol. 271, no. 3, pp. 239–254, 2018.

[5] C. Tian et al., “Deep learning on image denoising: An overview,” Neural Networks, vol. 131, pp. 251–275, 2020.

[6] M. Takeda, H. Ina, and S. Kobayashi, “Fourier-transform method of fringe-pattern analysis for computer-based topography and interferometry,” JosA, vol. 72, no. 1, pp. 156–160, 1982.

[7] Y. Aoyama, Y.-G. Park, C.-W. Lee, and D. Shindo, “Precise evaluation of charging effect on sio2 particles by simulation of holograms,” Materials transactions, vol. 43, no. 3, pp. 474–477, 2002.

[8] S. L. Y. Chang et al. "Performance of a direct detection camera for off-axis electron holography." Ultramicroscopy, vol. 161, pp. 90-97, 2016.

Author Biography

Takuma Okada received his BE in electronic engineering from Osaka University, Suita, Japan. He is a graduate student at the Graduate School of Information Science and Technology, Osaka University. His work has focused on the image processing for electron holograms.

Yoshihiro Midoh received his BE and ME in electronic engineering from Osaka University, Suita, Japan and his PhD in information science and technology from Osaka University. He is a specially appointed associate professor at the Graduate School of Information Science and Technology, Osaka University. His work has focused on the signal and image processing for advanced measuring equipment such as electron microscopy.

Koji Nakamae received his BE, ME, and PhD in electronic engineering from Osaka University, Suita, Japan. He is emeritus professors of the Graduate School of Information Science and Technology, Osaka University. His work has focused on CAD linked-electron beam test systems, signal and image processing, and the analysis of economics of VLSI manufacturing.

Noriyuki Miura received the BS, MS, and PhD in electrical engineering all from Keio University, Yokohama, Japan. He is a professor at the Graduate School of Information Science and Technology, Osaka University, Suita, Japan. His work has focused on hardware security/safety and next-generation heterogeneous computing systems