We propose an experimental depth-sectioned imaging method based on annular dark-field scanning confocal electron microscopy (ADF-SCEM). Four-dimensional (4D) datasets, consisting of 2D probe images taken at every probe position during 2D raster scanning, were acquired with an aberration-corrected scanning transmission electron microscope. A series of depth-sectioned images were constructed by processing a single 4D dataset. A pinhole and a stage-scan system used in earlier studies were not required in the present data acquisition. Optimal observation conditions for the 4D data acquisition in the ADF-SCEM mode are outlined from multislice simulations. [DOI: 10.1380/ejssnt.2018.247]

Keywords: Four dimensional scanning transmission electron microscopy (4D-STEM); Scanning confocal electron microscopy (SCEM); Annular dark-field (ADF); Depth sectioning; Multislice simulation

I. INTRODUCTION

Four-dimensional scanning transmission electron microscopy (4D-STEM) is a technique where two-dimensional (2D) convergent beam electron diffraction (CBED) patterns are recorded at each pixel of a 2D raster scan across a specimen. 4D-STEM can deliver much more structural information [1, 2] than conventional STEM where only integrated electron intensities are acquired. The 4D dataset of CBED patterns can be utilized for structural analysis such as ptychographic reconstruction [3–9], strain mapping [10–12], electric and magnetic fields imaging using differential phase contrast [13, 14], and composition and thickness measurements [15] with position-averaged CBED (PACBED) [16].

In this article, we demonstrate depth-sectioned imaging of a specimen using a 4D dataset acquired with annular dark-field scanning confocal electron microscopy (ADF-SCEM). Figure 1 schematically illustrates ADF-SCEM optics, where a focused electron probe is raster-scanned over the specimen as in conventional STEM. Electrons that pass through the specimen are re-focused using the imaging lens located below the specimen. An annular aperture is inserted to collect the electrons that scattered within the specimen. The re-focused probe images in real space are recorded at each probe position, instead of CBED patterns in reciprocal space. ADF-SCEM observations can be performed even with a TEM equipped with aberration correctors in both illuminating and imaging systems [17]. In earlier studies [18–35], a pinhole was placed above the detector to block the electrons that were scattered under out-of-focus conditions. Besides, a stage-scan system has been developed to move the specimen in a scanning manner while the electron probe was fixed, which enabled 3D imaging with a lateral (XY) atomic resolution [36, 37]. However, when 4D data acquisition is performed with ADF-SCEM, the pinhole and stage-scan system are not needed.

In the following sections, we describe how depth positions in the specimen can be revealed from a probe image acquired with ADF-SCEM and demonstrate the construction of depth-sectioned images from a single 4D dataset.

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FIG. 1. Schematic diagram of the optics of annular dark-field scanning confocal electron microscopy (ADF-SCEM).
II. MECHANISM OF DEPTH-SECTIONED IMAGING

The geometric optics diagram of ADF-SCEM in Fig. 1 suggests that only electrons scattered from the in-focus depth are re-focused at the center of recorded image. The depth position in the specimen can be associated with the image by the following equation:

\[ R = M \beta z \]  

(1)

Here, \( R \) denotes the radius from the image center, \( M \) the magnification in the imaging optics, \( \beta \) the scattering angle, and \( z \) the depth in the specimen. Different depths can be related to concentric circles with different \( R \) values, and thus a series of depth-sectioned images can be constructed.

In practice, \( R \), \( \beta \), and hence \( z \) in Eq. (1) span certain ranges. For instance, when the inner and outer radii in the image are taken as \( R_1 \) and \( R_2 \), and the inner and outer scattering angles selected by the annular aperture are \( \beta_1 \) and \( \beta_2 \), electrons that originated from depths \( z_1 = R_1/M\beta_1 \) to \( z_2 = R_2/M\beta_1 \) are regarded as signal.

Equation (1) suggests that larger \( M \) leads to a better depth resolution. However, for an overly large \( M \) the center of the probe image may not be detected with a CCD camera, depending on the position of the scanning electron probe. Hence, an optimal \( M \) value should be selected.

III. EXPERIMENTAL CONSTRUCTION OF DEPTH-SECTIONED IMAGES

Cobalt nanoparticles embedded in disordered carbon were deposited on a carbon film and used as a sample. 4D datasets were acquired under ADF-SCEM conditions using a JEOL JEM-ARM200F TEM equipped with third-order aberration correctors in both illuminating and imaging systems. The acceleration voltage was set at 200 kV. An annular aperture fabricated using focused ion beam machining was inserted at the objective aperture position. Probe images in real space were recorded using the diffraction imaging function of Gatan Micrograph Suite (GMS) software with an Orius200D CCD camera. This is in contrast with most other 4D-STEM experiments, where diffraction patterns in reciprocal space are recorded.

The recorded 4D datasets were processed using custom Gatan DigitalMicrograph scripts. Since the center of the probe images deviated with scanning, the probe images were aligned to a common center by means of cross correlation followed by manual fine adjustments. Subsequently, concentric masks were applied to the electron images. Figure 2(a) shows an example of the probe image recorded with a pixel size of 0.38 nm; the number of pixels of the image was 256 x 256 and only the central region of the image is displayed. Figure 2(b) illustrates five concentric masks applied. Electron intensities of pixels within each mask are summed to yield the ADF-SCEM signal at a given probe position. A series of depth-sectioned 4D-SCEM images is then constructed by performing this process at every probe position.

Figure 3 shows conventional ADF-STEM images taken with a probe-forming convergence semi-angle of 21.8 mrad and three different defocus values, which bring into focus Co particles located at different depths. Three Co particles under the in-focus conditions are designated as A, B, and C in Fig. 3. While a similar depth-sectioning technique has been reported in the literature [38–54], we note that all the Co particles in the view field are observed irrespective of the used defocus values.
FIG. 4. 4D-SCEM images constructed from a 4D dataset recorded with ADF-SCEM optics. The approximate inner and outer radii ($R$) of the applied masks are indicated for each image along with the corresponding depths ($z$) estimated using Eq. (1) and the respective inner and outer collection semi-angles of 26.0 mrad and 34.8 mrad.

Figure 4 presents 4D-SCEM images constructed from a 4D dataset that was recorded with ADF-SCEM from the same area as in Fig. 3. Here we used a small convergence semi-angle of 7.2 mrad and a scattering angle ranging from 26.0 mrad to 34.8 mrad. The magnification in the imaging lens optics was set at 400,000. The probe scanning was performed at 72 × 52 sampling and a spatial step of 1.2 nm. Electron images were recorded with 256 × 256 pixels and a pixel size of 0.38 nm. Consequently, the 4D dataset consisted of 72 × 52 × 256 × 256 pixels. The exposure time was set at 50 ms for each pixel. The total acquisition time was about 720 s, including readout time. Drift correction was not performed. The approximate inner and outer radii ($R$) of the applied masks are indicated for each image in Fig. 4. The corresponding depths ($z$) were calculated using Eq. (1) with the respective inner and outer collection semi-angles of 26.0 mrad and 34.8 mrad. Figure 4 demonstrates that Co particles existing at different depths are highlighted depending on the used values of $R$ and $z$. Note that the focused cobalt particles (A, B, and C) in the ADF-STEM images in Fig. 3 are observed in separate 4D-SCEM images in Fig. 4, each displaying a different depth section. These results demonstrate that a series of depth-sectioned images can be obtained from a single 4D dataset recorded in the ADF-SCEM configuration.

In the present study, a slow-scan CCD camera has been used to record electron images, so that the data acquisition time was as long as about 720 s. However, this time could be reduced by an order of magnitude using a direct electron detector with a superior readout rate [55]. Therefore, depth-sectioned images could be constructed within minutes, including the time for online data processing.

Eq. (1) implies that the use of a small range of the scattering angle ($\Delta \beta = \beta_2 - \beta_1$) can improve the depth resolution. We used $\Delta \beta = 34.8 - 26.0 = 8.8$ mrad, and the resulting 4D-SCEM signals were rather strong. A smaller range of the scattering angle can be adopted in a future work to enhance the depth resolution.

IV. MULTISLICE SIMULATIONS OF 4D-SCEM IMAGES

Optimal conditions for the 4D data acquisition in the ADF-SCEM mode were investigated using a multislice method [56]. We have developed a computational program for multislice simulations that employed elastic scattering factors parameterized up to a scattering vector of 6.0 Å⁻¹ by Peng et al. [57].

The simulation cell had dimensions of 4 × 4 nm along X and Y axes and a thickness of 30 nm along the optic...
FIG. 6. 4D-SCEM images simulated using a convergence semi-angle of 10 mrad and a scattering angle range of (a) 22–32 mrad and (b) 62–72 mrad.

FIG. 7. 4D-SCEM images simulated using a convergence semi-angle of 20 mrad and a scattering angle range of (a) 22–32 mrad and (b) 62–72 mrad.

axis (Z axis). Figure 5 depicts the simulation cell viewed along the Z axis. Five Co atoms were arranged at different depths of 5, 10, 15, 20, and 25 nm in the cell. To reduce the computational time, only the shaded area in Fig. 5 was scanned. In calculations, the simulation cell was divided into 0.5-nm-thick slices. X × Y sampling for both real space and reciprocal space was 512 × 512 pixels.

The acceleration voltage was set at 200 kV and the convergence semi-angle to 10 mrad or 20 mrad. The scattering angle spanned 22–32 mrad and 62–72 mrad. The electron probe was focused at the entrance surface of the simulation cell. Aberrations except defocus were ignored.

Similarly to the experiments, probe images were calculated. The electron intensities within concentric masks were summed for every probe position to construct 4D-SCEM images. Figure 6 shows the images simulated using a convergence semi-angle of 10 mrad and scattering angle ranges of (a) 22–32 mrad and (b) 62–72 mrad. The used R value and the corresponding z calculated with Eq. (1) are indicated for each image. When the scattering angle of 22–32 mrad is adopted, Co atoms located at different depths are observed simultaneously. Note that the images for R of 0.8–1.2 nm in Figs. 6(a) and 7(a) do not reflect the true depths of Co atoms because the range of z (25–55 nm) exceeds the thickness of the simulation cell (30 nm). By contrast, when a large scattering angle of 62–72 mrad is applied in Fig. 6(b), the five Co atoms are observed separately in a depth range consistent with the depth of the Co atoms. Thus, a high depth resolution can be achieved by selecting a large scattering angle.

In real situations, geometric aberrations of the imaging lens might deteriorate the depth resolution in 4D-SCEM if a large scattering angle of 62–72 mrad is selected. However, with newly developed aberration correctors the angle of uniform phase for an imaging system can be extended up to ~70 mrad [58, 59]. With this type of aberration correctors, 4D-SCEM is likely to maintain a high depth resolution even when a large scattering angle is adopted.

Next, we present the effect of convergence semi-angle on 4D-SCEM imaging. Figure 7 shows 4D-SCEM images simulated using a convergence semi-angle of 20 mrad and a scattering angle range of (a) 22–32 mrad and (b) 62–72 mrad. As compared with Fig. 6(a, b), the Co atom located at a depth of 5 nm in the simulation cell shows a strong contrast, while deeper Co atoms are blurred. These features can be attributed to the shape of the electron probe generated with a large convergence semi-angle. Figures 8(a) and 8(b) illustrate the probe intensity in an XZ (vertical) plane calculated using a convergence semi-angle of 10 mrad and 20 mrad, respectively. Figure 8(b) indicates that with the convergence semi-angle of 20 mrad, the electron probe has a sharp intensity peak at the focused depth. Hence the Co atom at a depth of 5 nm is illuminated with a high intensity, while the Co atoms at the lower depths are illuminated by a spread beam causing the image blur. By contrast, Fig. 8(a) shows that the electron probe produced with a convergence semi-angle of 10 mrad maintains a narrow shape over the entire thickness of the simulation cell (30 nm). Thus the use of a small convergence semi-angle should reduce the contrast near the entrance surface and the image blur at the lower depths. These results imply that the convergence semi-angle of 7.2 mrad used in our experimental study was appropriate for the 4D data acquisition.

V. CONCLUSION

Using ADF-SCEM we have acquired 4D datasets composed of the re-focused probe images taken at every probe

FIG. 8. Probe intensity in an XZ (vertical) plane simulated using a convergence semi-angle of (a) 10 mrad and (b) 20 mrad.

250 J-Stage: https://www.jstage.jst.go.jp/browse/ejssnt/
position during 2D raster scanning. We suggest that the radial distance from the center of the probe image can be associated with the depth position in the specimen. The depth-sectioning capability was confirmed experimentally by generating depth-sectioned images from a single 4D dataset. The use of a small convergence semi-angle used in the experimental observations was validated by multislice simulations. Processing of the 4D dataset can be performed online using GMS software within minutes after the data acquisition. Hence the presented 4D-SCEM method can be utilized as an efficient technique for 3D imaging.

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