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Three-Dimensional Analysis of Nanoparticles using Annular Dark-Field Scanning Confocal Electron Microscopy Established in a Double Aberration-Corrected Microscope

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Recently, the development of aberration correctors has contributed to the great improvement in the spatial resolution and analytical sensitivity in scanning transmission electron microscopy (STEM). In addition, aberration correction allows a large numerical aperture of an electron lens, resulting in a shallow depth of field of an image. Therefore, aberration-corrected STEM has been one of promising three-dimensional (3D) imaging techniques. In practice, Borisevich et al. demonstrated the 3D reconstruction of catalytic nanoparticles on a substrate particle using aberration-corrected high angle annular dark field (HAADF) STEM [1]. However, D'Alfonso et al. suggested that this incoherent STEM imaging suffers a "missing cone" problem in the 3D optical transfer function. As a result, the depth resolution is deteriorated for laterally extended objects [2]. Nellist et al. established an electron trajectry for scanning confocal electron microscopy (SCEM) in a double aberration-corrected microscope [3], which can be considered as an electron-optical version of scanning confocal optical microscopy. Compared to STEM, SCEM can dramatically improve the depth resolution due to rejecting electrons from an out-of-focal plane by a pinhole aperture at a conjugate plane. In addition, incoherent SCEM does not suffer the "missing cone" problem. Hashimoto et al. succeeded in the 3D reconstruction of carbon nanostructures using annular dark filed (ADF) SCEM established in a conventional TEM/STEM instrument (Cs=1 mm) [4]. However, the objects were apparently elongated with about 100 nm along the optical axis because of the insufficient depth resolution depending on the probe size. In this study, we established an ADF-SCEM trajectory in the double aberration-corrected microscope and observed nanoparticles on support materials. The 3D structure of the complex samples was analyzed and compared with the results obtained in an aberration-corrected STEM.

Figures 1(a) and (b) show schematic illustrations of STEM and ADF-SCEM trajectories with a stage-scanning system, respectively. The instrument we used was

Oxford-JEOL 2200MCO fitted with two aberration correctors, which was installed with a designed pinhole and an annular aperture. The stage-scannig system developed by Takeguchi et al. [5] allows a specimen to move in XYZ directions independently from the lens configuration. On STEM and SCEM, the condenser lens forms a fine probe in the specimen in the upper optics. Whereas in the lower, SCEM has the imaging lens excited to project and magnify the image of the probe on the pinhole plane. Detected signals are displayed on a computer screen synchronizing with the specimen displacement.

Figures 2(a) and (b) are aberration-corrected HAADF-STEM and ADF-SCEM XZ slice images of Au nanoparticles on an amorphous carbon film, respectively. The vertical XZ slices show the elongations of the nanoparticles along the optical axis. The elongations in both modes are much smaller than that taken by using a conventional microscope, which indicates the improvement in the depth resolution. However, the elongation on HAADF-STEM depends on the lateral particle size, while such dependence is not seen on ADF-SCEM. Furthermore, in the ADF-SCEM image, the carbon film can be observed as well as the nanoparticles. Therefore, the aberration-corrected ADF-SCEM is useful for the analysis of nanoparticles on support materials.

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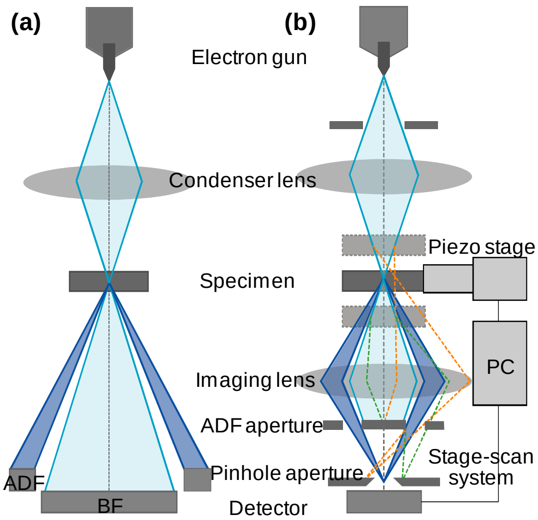

FIG.1. Schematic illustration of (a) STEM and (b) ADF-SCEM trajectories with a stage-scanning system.

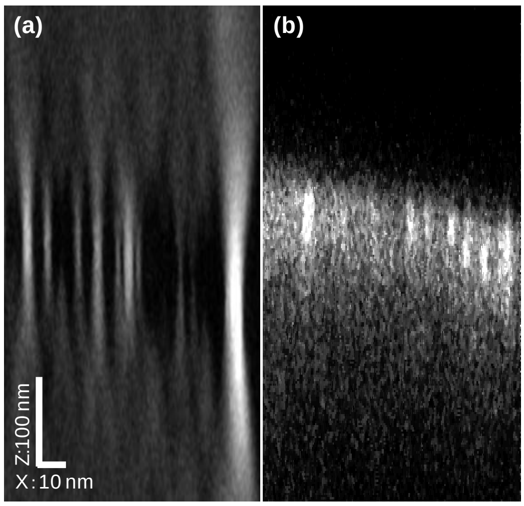

FIG.2 (a) Aberration-corrected HAADF-STEM and (b) ADF-SCEM XZ slice images of Au nanoparticles on an amorphous carbon film.