Mesoscopic Structure Analysis of Metallic Materials by Coherent X-ray Diffraction Microscopy

Coherent X-ray diffraction microscopy [1] (CXDM) is a novel technique for reconstructing the electron density of a sample. Figure 1 shows a schematic illustration of CXDM. By illuminating an isolated sample with coherent X-rays through an aperture, one observes its Fraunhofer diffraction patterns. The finely sampled diffraction intensities are measured by a two-dimensional X-ray detector. The phases of scattered X-rays are retrieved using an iterative algorithm, e.g., Fienup's hybrid input-output (HIO) algorithm [2], allowing the density of the sample to be reconstructed in two- or three-dimensions (2D or 3D).

CXDM has great potential as a technique for structural studies of metallic materials because it can be applied to micron-thick samples and is nondestructive. At present, the best image resolutions that have been achieved are 7 nm for a 2D image and less than 50 nm for a 3D image. The resolutions of transmission electron microscopy (TEM) and 3D atom probe (3DAP) microscopy are currently better than that of CXDM, but a TEM specimen must be less than 0.5 μm in thickness, while 3DAP destroys the specimen during the measurement. CXDM provides mesoscopic structure information about a micrometer sample. For example, precipitates in some aluminum alloys in practical use are a few tens of nanometers in size. CXDM is thus perfectly suited for the study of the exact 3D configuration of precipitates in a micrometer alloy sample. In the present study, the validity of CXDM for this kind of observation is demonstrated with an age-hardened aluminum alloy in practical use [3].

The chemical composition of the aluminum alloy was 96.376 at. % Al, 0.763 at. % Si, 0.010 at. % Fe, 1.887 at. % Cu, 0.431 at. % Mn, 0.515 at. % Mg, <0.005 at. % Cr, 0.004 at. % Zn, 0.006 at. % Ti, and <0.003 at. % Zr. Particles of the alloy of about 1 μm diameter were prepared and mounted on a 30-nm-thick Si_3N_4 membrane. A well-isolated particle was selected by optical microscopy. The CXDM measurement was carried out at beamline **BL29XUL** [4]. The membrane with the aluminum alloy particles was placed in vacuum. Incident X-rays of 5 keV irradiated the selected particle through a 20 μm aperture. Forward X-ray diffraction was collected by a charge-couple device detector placed 1.32 m downstream of the sample. Diffraction profiles were measured as a function of the sample rotation angle (α) around the horizontal axis perpendicular to the incident direction. The α value was changed from –70° to 70° in 5° steps. Each step took 2 hours.

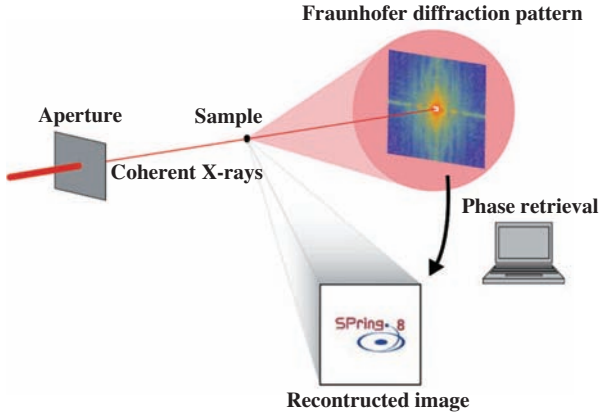

Fig. 1. Schematic illustration of coherent X-ray diffraction microscopy.

Figure 2(a) shows the diffraction pattern at $\alpha = 0°$. The fine structures in the diffraction pattern include information about both the shape of the sample and its internal structure. The sample image was reconstructed by the iterative normalization algorithm [5], which is a kind of HIO algorithm. Figure 2(b) shows the image reconstructed from the diffraction pattern shown in Fig. 2(a). The pixel size is 20.4 nm in both x and y directions. The sample image is displayed in gray scale, with darker colors corresponding to higher electron densities. In this image, the contrast results not only from variations of the sample thickness but also from the distribution of precipitates in the sample. To clarify the internal structure, a 3D image was reconstructed. The 29 diffraction patterns at different α values were normalized by the sum of the 2D reconstructed intensities at each α. A 3D diffraction intensity distribution with $561 \times 561 \times 561$ pixel resolution was made from the normalized 2D diffraction patterns shown in Fig. 3(a). Figure 3(b) shows the 3D reconstructed image. The dark blue area shown in Fig. 3(b) represents the surface figure of the reconstructed 3D image. The voxel size is 29.1 nm on each side. The particle size is about $1280 \times 850 \times 1080$ nm^3. According to Silcock [6], the *S*-phase, which has the composition of Al_2CuMg, precipitates in this aluminum alloy. Thus, we identified the high-electron-density regions resulting from *S*-phase precipitates based on the electron densities of Al and Al_2CuMg calculated from their atomic structures, which are 7.8×10^2 and 1.0×10^3 electrons/nm^3, respectively. Assuming that the voxel with a maximum value in the 3D reconstruction corresponds to pure Al_2CuMg, we trace the region with more than

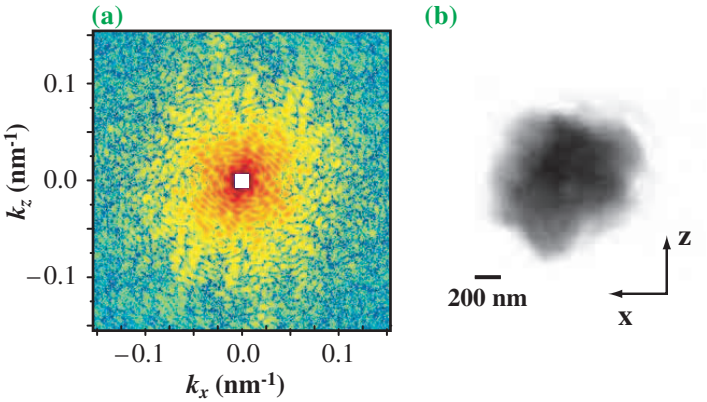

Fig. 2. **(a)** Coherent X-ray diffraction pattern of the Al alloy particle in 801×801 pixels at α=0°. The region of the central 37×37 pixels is an unmeasurable area due to the direct beam stop. **(b)** Image reconstructed from the diffraction pattern of **(a)**. The pixel size is 20.4 nm.

77% of the maximum density, corresponding to the border of the precipitates. The surface image of the high-electron-density region is drawn inside the particle image in Fig. 3(b). The high-electron-density region is about 780×480×220 nm^3 in size.

In conclusion, the internal structure and the shape of a micrometer-sized sample of a precipitation-hardened aluminum alloy were visualized by three-dimensional CXDM, and the spatial distribution of *S*-phase precipitates was evaluated. To examine the shape of each nanometer-sized precipitate, or the interface between the Al and the precipitates, it is necessary to improve the resolution of CXDM by using more brilliant X-rays. This will be realized using a next-generation light source such as an X-ray free electron laser (XFEL). By using an XFEL, *in situ* observation during heating might be realized with nanometer resolution.

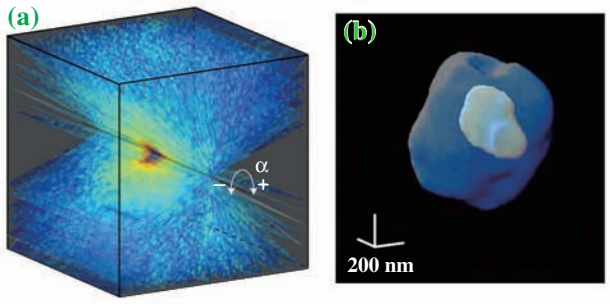

Fig. 3. **(a)** Three-dimensional diffraction array of 561×561×561 pixels made from 29 diffraction patterns of the Al alloy particle within –70° ≤ α ≤ 70° with 5° steps. **(b)** Three-dimensional surface figure of the Al alloy particle reconstructed from the diffraction array. A high-electron-density region resulting from Al_2CuMg precipitates, which is derived using the known electron density, is drawn inside the particle image.

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