1207 – Improving the Depth Resolution of HAADF Sectioning by 3D Deconvolution

Akimitsu Ishizuka[1], Kazuo Ishizuka[1], Ryo Ishikawa[2], Naoya Shibata[2], Yuichi Ikuhara[2], Hiroki Hashiguchi[3] and Ryusuke Sagawa[3]

[1]HREM Research Inc., Higashimatsuyama, Saitama, Japan, [2]University of Tokyo, Bunkyo-ku, Tokyo, Japan, [3]JEOL Ltd., Akishima, Tokyo, Japan

In a microscope the lateral resolution and the depth resolution have the following relationships: $0.6\lambda/\sin\alpha$ and $\lambda/\sin^2\alpha$, respectively. Here, α is the objective aperture angle for a wide field microscope, and the probe formimg angle for a scanning microscope. Since the optical microscope has a large image forming angle, we can successfully observe a cross section of the sample in an order of a wavelength. In a scanning transmission electron microscope (STEM), with the advent of Cs correction technology a convergence angle for probe formation can be increased substantially. Then, the possibility of observing the cross section of a sample has been discussed [1]. However, contrary to an optical microscope the convergence angle is still only several tens of mrad, and the ratio of the depth resolution to the lateral resolution is rather large, namely $1/0.6\alpha$. For example, Ishikawa et al. reported an expected depth resolution < 1.0 *nm* for $\alpha = 100$ mrad at an acceleration voltage of 300 kV [2]. However, the depth resolution is far inferior to the lateral resolution. In this report, therefore, we have developed the technique for 3D HAADF image deconvolution to improve the depth resolution.

When we acquire a set of through-focus HAADF images (a 3D HAADF image stack), each image may be represented by a convolution of a 3D scattering distribution and the 3D probe for a correspond-ing defocus. Namely, a 3D HAADF image stack is given by a convolution of a 3D scattering distribution and a 3D probe. Then, in principle, the 3D scattering distribution can be estimated from a 3D HAADF image stack by deconvoluting with a 3D probe. However, the 3D HAADF probe extends infinitely in the z direction as shown in Figure 1a. This is because every section of the 3D HAADF probe is normalized to one owing to the conservation of propagating electrons. Therefore, when the sample is a uniform slab, the HAADF signal does not become negligible even for an infinite defocus. In other words, the 3D HAADF probe is not band limited in terms of Fourier analysis along the beam propagation direction. For this reason, a simple deconvolution technique using a 3D Fourier transform does not work.

For a (2D) HAADF-STEM image we have developed the deconvolution routine in order to improve the lateral resolution based on the Richardson-Lucy algorithm and the maximum entropy method [3]. Here, we assume that the sample is thinner than the depth of focus (depth resolution), and that the HAADF signal can be represented by the 2D convolution between the projection of scattering distribution and the 2D probe. Since the size of the 2D probe is an order of atomic image, deconvolution can be performed ideally using the 2D Fast Fourier Transform (FFT) except the image edge. However, we cannot simply extend the FFT technique for a 2D HAADF image to a 3D HAADF image stack, since the 3D HAADF probe is not band limited along the z direction. Furthermore, due to the infinite nature of the 3D HAADF probe, a regular way of applying the Richardson-Lucy algorithm [4] does not work.

Therefore, we have developed a unique deconvolution method for a 3D HAADF image stack based on the maximum entropy method. The new method requires an observation of a limited range of through-focus HAADF images, that covers only the sample as well as some vacuum regions. In the method, we do not directly deconvolute the observed 3D HAADF image by the 3D probe using the 3D FFT, but convolute the 3D probe with the estimation function that will represent the scattering distribution. The estimation function is iteratively adjusted so that the convolution of the estimation function and the 3D

probe reproduces the observed data. Here, a 3D convolution is performed by a 2D convolution over each image plane using the 2D FFT, and by a convolution along the *z*-direction in real space.

Figure 1 illustrates the case of deconvolution of a single object of Gaussian. The convergence semi-angle is 60 mrad for 300kV electrons. The standard deviation of the Gaussian is 0.07 nm. Due to the cone-shape of the prove (a), the width of the convoluted image is elongated to about 4 nm as shown by the blue line in (b) along the z-direction. This width becomes to 1.3 nm, about one-third of the original, after 1000 iterations as shown by the red line in (b).

Figure 2 shows a more realistic case of heavy atom dopants. The convergence semi-angle is 60 mrad for 300kV electrons. (a) shows the probe convoluted with the Gaussian (FWHM=0.06 nm) to include a physical probe size effect. The line-profile of the simulated through-focus HAADF image of three Ce dopants in *w*-AlN [5] is shown by the blue line in (b), while the line profile after deconvolution is shown by the red line. The Ce peaks become sharp, and its width is reduced to about one half after 1000 iterations.

In the presentation, we will show the results applied to the experimental images.
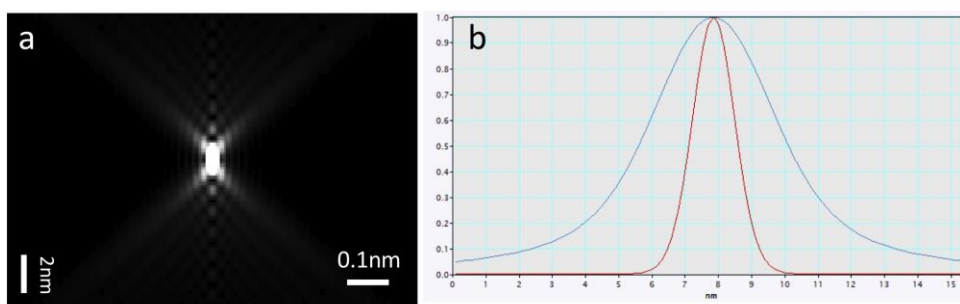

Figure 1. Deconvolution of a single object of Gaussian. (a) xz-section of the 3D probe. (b) line-profiles of the convoluted Gaussian and the 3D probe along the z-direction in blue, and of the deconvolution after 1000 iterations in red.
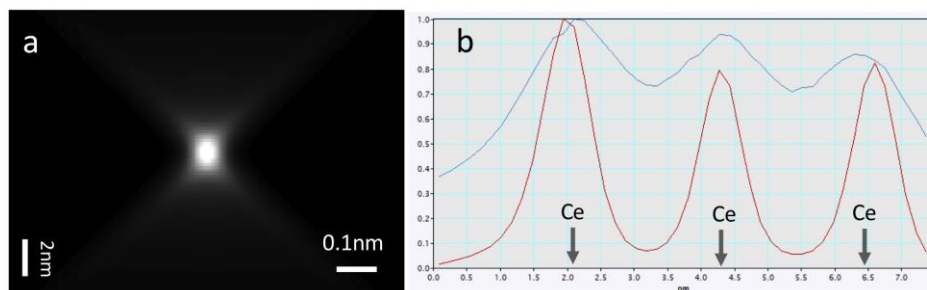

Figure 2. Deconvolution of the simulated 3D HAADF image of three Ce dopants in w-AlN. (a) xz-section of the 3D probe. (b) Line-profile of the simulated through-focus HAADF image of three Ce atoms in blue, and of the deconvolution after 1000 iterations in red. The arrows show the Ce positions in the mocel.

References
[1] A.Y. Borisevich, A.R. Lupini, S.J. Pennycook (2006) *Proc. Natl. Acad. Sci.* **103**, 3044.
[2] R. Ishikawa, A.R. Lupini, Y. Hinuma, S.J. Pennycook (2015) *Ultramicroscopy* **151**, 122.
[3] DeConvHAADF for DigitalMicrograph: www.hremresearch.com/Eng/plugin/HAADFEng.html
[4] W.H. Richardson (1972) *J. Opt. Soc. Am.* **62**, 55; L.B. Lucy (1974) *Astron. J.* **79**, 745.
[5] R. Ishikawa, S.J. Pennycook, A.R. Lupini, et al. (2016) *Appl. Phys. Lett.* **109**, 163102.