

## Controlling Depth Resolution of Phase Images by Ptychography using Achromatic Condition

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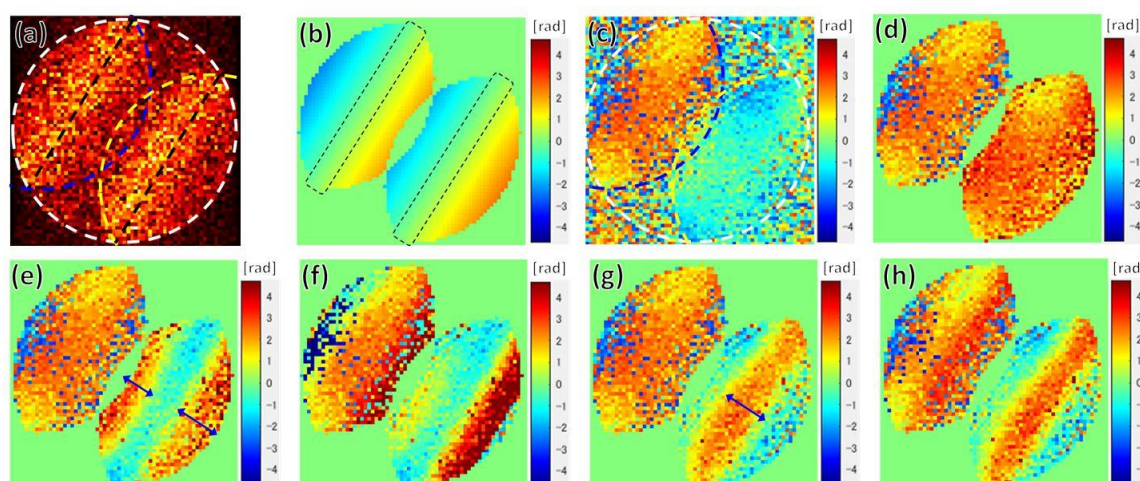
Owing to the development of aberration correctors in electron microscopy, lateral resolution (X-Y directions) has been improved down to ~40 pm in scanning transmission electron microscopy (STEM) [1]. Meanwhile the resolution in depth (Z) direction has recently gained much interests as it is important when mapping the whole 3-dimensional (3D) structure of a specimen [2]. It is effective to use a beam with larger convergence angle to enhance the depth resolution ( $d_z$ ), since  $d_z$  have a relation with the convergence angle ( $\alpha$ ) and wavelength of the electrons ( $\lambda$ ), as  $d_z = \lambda/\sin^2(\alpha)$ . Further correction of higher order aberrations, the effective convergence angle improves as we can use a large condenser lens aperture of up to 70 mrad semi-angle, resulting in a few nm of depth resolution at 300 kV in High Angle Annular Dark-Field (HAADF)-STEM images. Although the method using multiple HAADF-STEM images is popular to obtain depth resolution, there is another way to use more advanced detectors to utilize the rich specimen information on the detector plane. Recently, pixelated STEM detectors with fast frame rate of several thousand frames per second and several ten thousand pixels per frame have been developed and commercialized [3,4]. Using the 4-dimensional (4D) dataset obtained by the detectors, one can reconstruct a phase image through an image processing method, that is, ptychography [5,6]. As the process includes phase manipulation so as to maximize the image contrast of specimen, phase images with high signal-to-noise (S/N) ratio are obtained. The technique to remove residual aberrations are also developed by adding aberration correction functions during the reconstruction process. This also enables the creation of through-focus images, which in turn, serve as the optical depth sectioning technique for reconstructing the 3-dimensional structure of specimen [7]. In this paper, we aim to prove the ability to control the depth resolution of the through-focus images by further manipulation of the signal on detector planes.

We have used a pixelated STEM detector (4DCanvas<sup>TM</sup>, JEOL,) with a direct electron CCD image sensor (pnCCD, PNDetector) whose maximum readout speed is 7,500 fps [3,4]. The confirmed operable accelerating voltage is 20 ~ 300 kV. For experiments in this paper, the detector was integrated into an aberration corrected 200 kV electron microscope (NEOARM, JEOL). As this detector is installed below the conventional ADF detector, we can simultaneously record the 4D dataset and the HAADF-STEM image. In our experiments, the data was taken with a frame rate of 7,500 fps (~133  $\mu$ s in dwell time per pixel). The fast readout enabled recording of 256 x 256 pixels scan within 10 seconds.

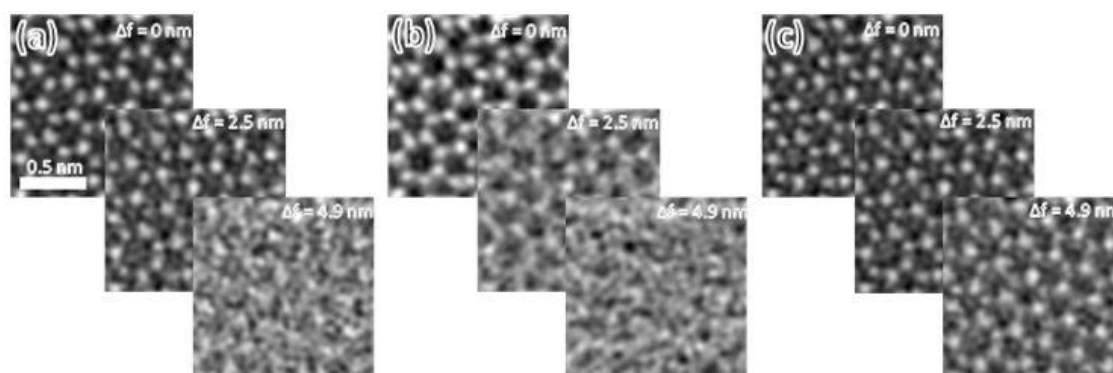
In the ptychography, the signal integration areas (shape and position) are dynamically changed for corresponding spatial frequencies. If we look at the interference signal between transmitted and diffracted beams, one can notice that the signal amplitude has its maxima at certain areas (Fig. 1(a)). The areas are called achromatic lines, as the phase in the areas is not affected by the fluctuation of electron beam in depth direction due to distribution of wavelengths of electrons. This could be due to the instability in the electric current of the objective lens, and so on. This effect is obvious if we map the phase shift due to defocus. The shift is almost zero only in the areas near the achromatic lines as shown in Fig. 1(b). In normal ptychography operation to obtain phase image with enhanced contrast, we simply add  $\pi$  to one of the two interference areas to make them in phase to enhance the phase image contrast as shown in Figs. 1(c) and (d). When we added  $\pi$  only to the other area than the achromatic condition to make the phase image contrast sensitive to defocus as shown in Figs. 1(e) and (f), we expect to enhance depth resolution.

We call this process as process A. Inversely, when  $\pi$  is added only to the area of achromatic areas as shown in Figs. 1(g) and (h), the image contrast is expected to be insensitive to defocus, resulting in longer focal depth. We call this process as process B. Figure 2 shows reconstructed through-focus phase images of mono-layer graphene at 80 kV by ptychography. Fig. 2(a) shows images with normal ptychography process. Fig. 2(b) shows images after the process A. And Fig. 2(c) shows images after the process B. Compared with the image series in (a), the series in (b) start to blur more quickly and the series in (c) keep the lattice image contrast even at 4.9 nm of defocus.

While the technique to improve the depth resolution introduced here can directly be used for the optical depth sectioning, the technique to make focal depth longer without losing spatial resolution in x-y directions can be used for tilted sample in tomography, where the blur in image due to defocus is undesirable.



**Figure 1.** Ptychographic signal at a certain spatial frequency of mono-layer graphene. (a) Amplitude map. Achromatic lines are indicated as black dashed lines. (b) Phase shift due to defocus. The shift is almost zero inside the area indicated by dashed rectangles. (c) Original phase map. (d) Phase map after adding  $\pi$  to one of the interference areas. Amplitude in the areas other than the interference areas are set to zero in order to remove unnecessary signal for denoising. (e) Phase map after adding  $\pi$  only to the area where achromatic condition is not met (indicated by arrows). (f) Phase map after adding 3 nm of defocus to (e). (g) Phase map after adding  $\pi$  only to the achromatic areas (indicated by an arrow). (h) Phase map after adding 3 nm of defocus to (g).



**Figure 2.** Reconstructed through-focus phase images by (a) normal ptychography process, (b) higher depth resolution method and (c) lower depth resolution method. Condenser lens aperture of 40 mrad in

semi-angle was used. The defocus value is indicated in each image. Compared with defocused images in (a), the images in (b) are more blurred and the lattice contrast is more preserved in the images in (c).

#### References

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