Elements of quantitative ADF imaging for crystallography; quantitative, precise and reproducible methodology

Kimoto, K.[1], Yamashita, S.[1], Kikkawa, J.[1] and Ishizuka, K.[2,1]

[1] National Institute for Materials Science, Japan, [2] HREM Research Inc., Japan

Annular dark-field (ADF) imaging [1] allows us to analyze crystal structures using incoherent imaging approximation. Quantitative ADF imaging has been reported [2], in which the ADF intensity is normalized by the incident probe intensity. We found the nonlinear response of conventional ADF imaging system becomes critical [3] in high sensitivity measurements, and we demonstrated a quantitative ADF imaging, in which the number of electrons at each pixel was evaluated. We also estimated effective source distribution using a high-resolution ADF image of a monolayer graphene (Fig. 1) [4]. Because the number of electrons was measured, the quantum noise of the ADF images was estimated, which became a guideline to assess the difference between experiment and simulation.

The precision of the ADF imaging suffers a low ADF signal. The ADF signal from a monolayer graphene is only several electrons per pixel under a typical experimental condition; e.g., probe current of 30 pA, ADF inner angle of 50 mrad, and 0.04 ms of dwell time. We observed multiple ADF images with short dwell time to minimize an image distortion due to image drifts [5]. We prepared DigitalMicrograph script to track the specimen drift during multiple image acquisitions, resulting in a high precision of several picometers in atomic position analysis [6]. Dynamical scattering or crystal tilt are problematic even in the case of ADF imaging [7], which suggests the limit of the incoherent imaging approximation.

The reproducibility of ADF imaging depends on various experimental parameters, including aberrations of the objective lens. The ADF image of a monolayer graphene shows small systematic deviation from the simulation, and we recognized the time-dependent low-order aberrations are a barrier for further quantitative analyses. We proposed a method to measure low-order aberrations using two Ronchigrams of unknown crystalline specimen [8]. Fourier transforms of two Ronchigrams observed under different foci were used to measure the aberration, such as defocus, two-fold astigmatism, coma and three-fold astigmatism. Figure 2 shows an example of aberration measurement, in which the high stability of defocus C_1 and two-fold astigmatism $\boldsymbol{A_1}$ of less than 1 nm per minute is demonstrated.

The recent progress in the instrumentation of 4D-STEM allows us to apply it for crystal structure analyses, which was a time-consuming approach in the conventional instrument [9]. The crystal structure analysis using ADF imaging becomes more practical using fast 4D-STEM instrumentation.

[1] S.J. Pennycook, D.E. Jesson, Ultramicroscopy **37** (1991) 14-38.

[2] J.M. LeBeau, et al., Physical Review B **80** (2009)

[3] S. Yamashita, et al., Microscopy **64** (2015) 143-150.

[4] S. Yamashita, et al., Microscopy **64** (2015) 409-418.

[5] K. Kimoto, et al., in: Proceedings of IMC16, Sapporo, 2006, p. 609.

[6] K. Kimoto, et al., Ultramicroscopy **110** (2010) 778-782.

[7] Y.G. So, K. Kimoto, J. Electron Microsc. **61** (2012) 207-215.

[8] K. Kimoto, K. Ishizuka, Ultramicroscopy **180** (2017) 59-65.

[9] K. Kimoto, K. Ishizuka, Ultramicroscopy **111** (2011) 1111-1116.

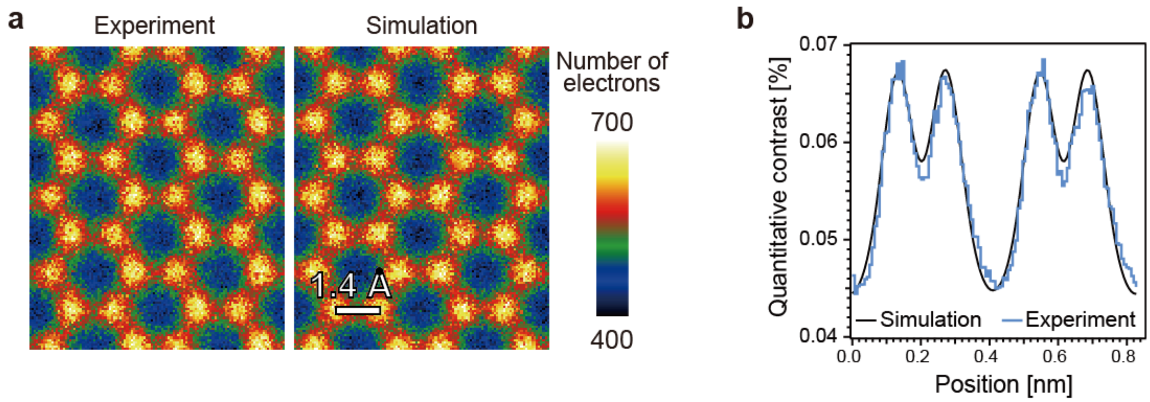

Fig. 1 a Quantitative ADF imaging of monolayer graphene. An experimental image was observed at 80 kV with a probe current of 27 pA. Multiple fast acquisition and drift correction were performed. Simulation includes residual aberrations, defocus spread and an effective source distribution, and the quantum noise is added on the basis of the Poisson process [4]. **b** Quantitative comparison between experiment and simulation that without the quantum noise.

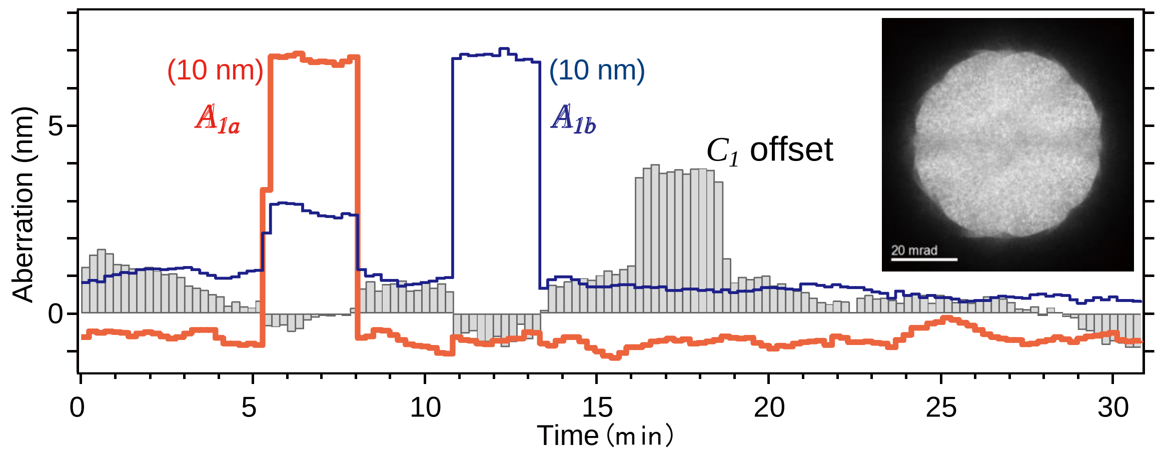

Fig 2. Time dependence of defocus C_1 and two-fold astigmatism $\boldsymbol{A}_1$ as measured using our method [8]. A specimen was a $SrTiO_3$ (001) film and the inset shows an experimental Ronchigram.

The authors would like to express their thanks to Dr. Shogo Koshiya, Dr. Naoki Ohashi and Prof. Katsuro Hayashi for invaluable discussions. This study was partly supported by a Grant-in-Aid for Scientific Research on Innovative Area "Mixed Anion" (Project JP16H06440) of JSPS. K.K. also thanks the MEXT Elements Science and Technology Project for support.