

## 20-kV Diffractive Imaging of Graphene by using an SEM-based Dedicated Microscope

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The optical microscope has reached a resolution finer than the wavelength of light; however, in the electron-microscopy field, decrease aberrations of lenses remains a challenge. Even with the help of aberration correctors and monochrometers, the resolution is limited to more than multiples of ten times the wavelength of the electron beam [1]. On the other hand, diffractive imaging, which is an imaging method using iterative phase retrieval from a diffraction pattern [2], can obtain high-resolution images without suffering any aberrations of the imaging lens. This method (using low acceleration voltage) have been applied by the authors to reach atomic resolution with a dedicated microscope based on a conventional scanning electron microscope (SEM) [3, 4], and this microscope resolved the atomic arrangement of a single-wall carbon nanotube at 30 kV [5]. In the present study, the atomic arrangement of multi-layer graphene in the case of an acceleration voltage of 20 kV was reconstructed, and the possibility of reconstruction of a non-periodic structure was investigated.

Diffraction patterns were experimentally recorded by using a dedicated microscope based on in-lens type SEM (S-5500, Hitachi High-Technologies Corporation) with a film-loader system of a transmission electron microscope [4]. Graphene films were synthesized by CVD and transferred on a specimen stage with fine hole of about-75-nm diameter.

A pattern of multi-layer graphene reconstructed from a diffraction pattern recorded by using an electron beam with an acceleration voltage of 20 kV (namely, wavelength of 0.00859 nm) is shown in Figure 1(a). Each bright spot (“atomic column” hereafter) shows the intensity of a single carbon atom or a few carbon atoms. This resolved atomic arrangement of graphene shows the resolution of this imaging is finer than 0.12 nm (14 times the wavelength, which is close to the results with aberration correctors). The atomic arrangement shown in Fig. 1(a) is composed of three different contrasts (low, medium, and high intensities) in each carbon atomic column. And the order of the contrast of the atomic column is regularly repeated. A model of the atomic arrangement of ABA-stacking tri-layer graphene is shown in Figure 1(b), and it shows three different forms of stacking carbon atoms (i.e., single atom, two atoms, and three atoms). Intensities of the pixels along lines #1 to #8 (open and close symbols), which correspond to each line in Fig. 1(a) and fitted Gaussian distribution are shown in Figure 1(c). Comparison the Gaussian distribution with the atomic arrangement model and a simulation of projected potential reveals that the graphene shown in Fig. 1(a) has tri-layer structure with ABA stacking.

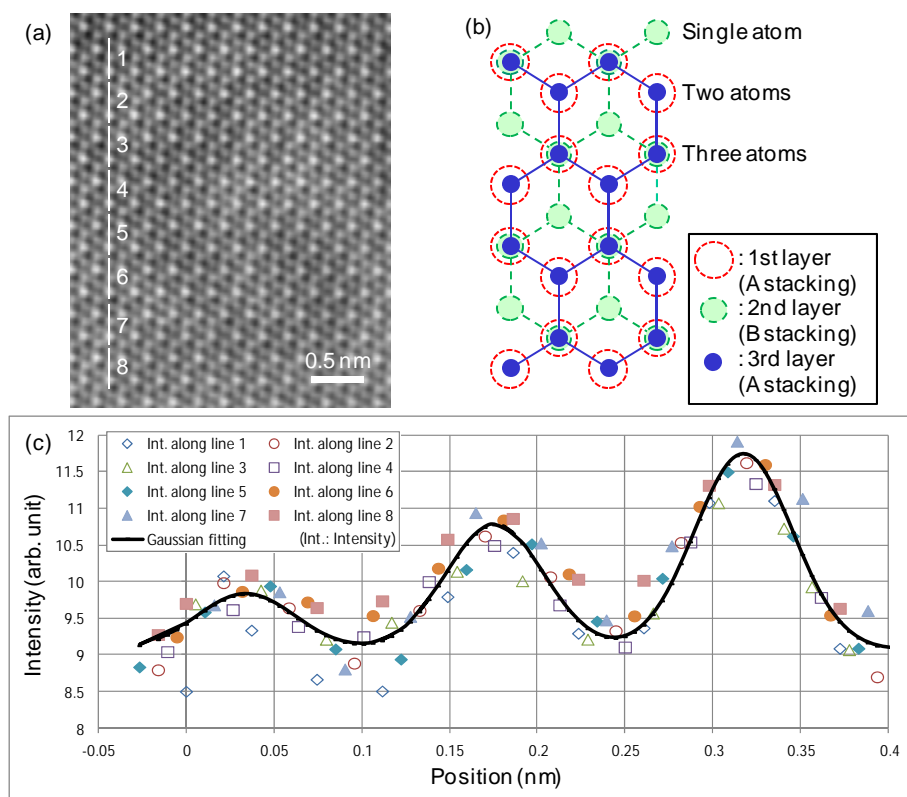
To investigate issues concerning reconstruction of a non-periodic structure such as grain boundaries or defects in crystals, diffraction patterns of defective graphene with various noises were simulated, and the atomic arrangement of the graphene was reconstructed from the simulated diffraction patterns. The defective structure could be reconstructed from a diffraction pattern with high signal-to-noise ratio, but only the periodic atomic arrangement of graphene was reconstructed from a low-signal-to-noise-ratio

diffraction pattern. It is thus concluded from this result that a defective atomic arrangement can be imaged by diffractive imaging and that high-signal-to-noise-ratio recording of diffraction patterns is essential to reconstruct a non-periodic structure.

In summary, low-voltage diffractive imaging was shown to have the possibility of atomic-resolution imaging that can reconstruct not only periodic structures but also non-periodic structures. It is concluded that this method will open up new possibilities of application of electron microscopy.

#### References:

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**Figure 1.** (a) A pattern of graphene reconstructed from a diffraction pattern obtained at 20 kV. (b) Atomic-arrangement model of ABA stacking tri-layer graphene. (c) Intensity distributions of reconstructed pattern along eight lines and fitted Gaussian distribution with three peaks.