Resolution Improvement in Aberration-Corrected Low-Voltage TEM with Monochromator at 60 kV

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Abstract. We have developed a low-voltage electron microscope equipped with a monochromator and Delta-type Cs correctors, which shows atomic resolution at accelerating voltages of 60, 30 and 15 kV. In theory, resolution of TEM images at 60 kV is severely affected by chromatic aberration, which is proven by our calculations of contrast transfer functions and multi-slice image simulation taking chromatic aberration into account with experimental conditions. Experimentally, TEM images of gold nano-particles were observed with non-monochromated and monochromated electron sources at 60 kV. Detectable spatial frequency in the image with the monochromated source was higher than that with non-monochromated source. We have demonstrated that the TEM image resolution at the low-voltage is improved by using a monochromated electron source, which reduces the energy spread of the electron source.

1. Introduction
Low-voltage transmission electron microscope (LVTEM) becomes essential tool to study nano-structure and chemical analysis of carbon related materials, since the LVTEM enables us to observe them with small radiation damage. In theory, the resolution in LVTEM is limited by diffraction limit ($d_d = 0.61 \lambda / \alpha$, where $\lambda$ and $\alpha$ are wavelength of electrons and acceptance angle for the imaging). The wavelength of electrons at 60 kV is 4.87 pm, which is approximately double of that for 200 kV, resulting in double diffraction limit with identical aperture angle. To improve the resolution at low accelerating voltage, reduction of the diffraction limit is mandatory. Therefore, we have developed a Delta-type aberration corrector that is composed of three dodecapoles [1]. It can correct fifth-order aberrations including six-fold astigmatism as well as third-order spherical aberration. With the corrector in probe forming system, atomic spacing of carbon-to-carbon in a graphene was clearly resolved in STEM images at 30 kV [2]. The Delta corrector can be applied to image forming lens system. Diffraction limit can be decreased by the large acceptance angle for the imaging realized by the corrector. Though we applied the Delta corrector for image forming lenses experimentally, the resolution of TEM images was not enough to detect the atomic structure of mono-layered materials [3]. It was concluded that the result was caused by a chromatic aberration, because TEM resolution is...
affected by defocus spread more severely than the resolution of a dark field STEM. Defocus spread caused by the chromatic aberration is defined as $C_c \frac{dE}{E_o}$, where $C_c$, $dE$ and $E_o$ are chromatic aberration coefficient, energy spread of electron source and primary electron energy. The defocus spread becomes larger when $E_o$ becomes smaller with constant $dE$. To improve the resolution in TEM images at lower accelerating voltages, correction of chromatic aberration $C_c$ of the image forming lenses or reduction of the energy spread $dE$ are required.

2. Experimental methods

In the present study, a newly developed LVTEM equipped with a monochromator and a Delta corrector for image forming lens systems was used. The microscope can work at accelerating voltage of 60, 30 and 15 kV. The accelerating voltage of 60 kV was used in this work. The monochromator consists of two Wien filters and an energy selection slit [4]. The energy width of the electron source can be changed by selecting slit width. The minimum energy width of the electron source was measured to be less than 25 meV with exposure time of 0.5 seconds. The coefficients of third-, fourth-, and fifth-order aberrations have been measured to be less than 1 μm, 30 μm, 0.1 mm, respectively. The fifth-order aberration of 0.1 mm induces $\pi/4$ phase shift in 57 mrad, which corresponds to the spatial frequency of higher than $(0.09 \text{ nm})^{-1}$. The measured chromatic aberration coefficient at 60 kV for the image forming system was 0.75 mm.

![Figure 1](image)

**Figure 1.** Calculated contrast transfer functions (blue) and envelope functions (green) at 200 kV ((a), (b) and (c)) and 60 kV ((d), (e) and (f)). Energy width of (a) and (d) is 0.8 eV, and that of (b) and (e) is 0.3 eV, and that of (c) and (f) is 0.1 eV. Chromatic aberration coefficient is 0.75 mm. Third-order spherical aberration coefficient and defocus are 10 μm and -6.1 nm at 200 kV and 3 μm and -4.7 nm at 60 kV, respectively.

3. Simulations

To evaluate the effect of the energy spread of an electron source on TEM images at lower accelerating voltage, we have calculated contrast transfer function (CTF) at two accelerating voltages. Figure 1 shows the CTFs at 200 and 60 kV with several energy spreads of the electron source. Chromatic aberration coefficient of 0.75 mm is used for the calculation. Here, an indicator of the spatial frequency for the information limit is defined as the frequency when intensity of the envelope function reduces to be 1/e (= 0.368). In the case of 200 kV, information limit of the envelope function is higher...
than \((0.1 \text{ nm})^{-1}\) even if the energy width is 0.8 eV, which is the typical energy width of a Schottkey source. On the other hand, in 60 kV, it is difficult to achieve \((0.1 \text{ nm})^{-1}\) with the energy width of even 0.3 eV which corresponds to the typical energy spread of a cold field-emission source. To achieve the information limit better than \((0.1 \text{ nm})^{-1}\) at 60 kV, monochromated source is required. This indicates that the reduction of the energy spread is important for improving the information limit at 60 kV.

Figure 2 shows simulated TEM images of a gold nano-particle from <111> direction at 60 kV by the multi-slice method. The effect of the defocus spread is included in the simulation by multiplying the Fourier transform of the exit wave by the envelope function due to the defocus spread. When the energy width is 0.8 eV, the contrast of atomic columns in the particle are blurred. By reducing the energy width to be 0.1 eV, the atomic columns are clearly resolved in the image. The simulation indicates the resolution enhancement using a monochromated source in TEM images at 60 kV.

![Figure 2](image)

**Figure 2.** Atomic model (a) and simulated TEM images of a gold nano-particle from <111> with the energy width of (b) 0.8 eV, and (c) 0.1 eV. Chromatic aberration coefficient of 0.75 mm and defocus of - 2.0 nm are used in the simulation.

### 4. Results & Discussion

Figures 3 show TEM images of gold particles and their power spectra of the Fourier transforms taken at 60 kV with the different energy spreads of the electrons. Non-monochromated electron source without energy slit and monochromated source with the slit width of 1.3 µm were chosen for the imaging conditions. The energy width of these electron sources were 0.77 eV and 0.10 eV, respectively. The atomic columns of the gold nano-particles were not clearly observed in the image with the non-monochromated source [Fig. 3(a)], though lattice fringes were detected in the image. On the other hand, the atomic columns of the gold nano-particles were detectable in the image with the monochromated source [Fig. 3(b)]. The power spectrum of the image from the non-monochromated source shows that the continuous bright contrast, which is originated by structural information of the amorphous carbon film, exhibits up to around \((0.2 \text{ nm})^{-1}\). The contrast in the power spectrum with the monochromated source extends to around \((0.1 \text{ nm})^{-1}\). Spots around \((0.1 \text{ nm})^{-1}\) from the gold-crystalline structure were detected in the image with monochromated source. Although these power spectra cannot be simply used for evaluating the resolution of the TEM images quantitatively because of non-linear contrast [5], these images and power spectra apparently show that the resolution of the TEM images is improved by using monochromated electron source.

The resolution of TEM images can be affected by both envelope functions due to spatial coherence and temporal coherence of the electron source, generally. Our monochromator produces achromatic and stigmatic illumination beam after the second Wien filter. Since a small cross-over for illumination makes a small illumination angle, the illumination angle for imaging is sufficiently small. And the aberrations up to sixth order were corrected by the image-forming aberration corrector. Thus, the effect of the envelope function due to the spatial coherence, which is caused by the illumination angle and the geometrical aberration, is sufficiently small. Therefore, the effect of the envelope function due to the spatial coherence can be neglected in the TEM images in Fig. 3. The resolution was dominantly affected by the envelope function due to temporal coherence in this observation.
We experimentally demonstrated that the resolution of the aberration-corrected TEM image was enhanced by the monochromated electron source. The experimental result indicates that the energy spread of the electron source is a dominant factor for information transfer at low accelerating of 60 kV [6], whereas the defocus spread from fluctuations of accelerating voltage and lens currents can affect the resolution in TEM.

Figure 3. TEM images of gold nano-particles taken at 60 kV with (a) non-monochromated source and (b) monochromated source. (c) and (d) are power spectra of the Fourier transform of (a) and (b), respectively.

5. Conclusion
We have developed the aberration-corrected LVTEM equipped with the monochromator, which shows the atomic resolution at 60 kV. The CTFs and the simulated TEM images show that the information limit can be improved by reducing the energy spread of the electron source. The TEM images of the gold nano-particles with non-monochromated and monochromated electron source, taken at 60 kV, showed that narrower energy spread of the electron source improved the resolution of TEM images.

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