Study of Point Spread in the Aberration-Corrected Transmission Electron Microscopy

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Abstract: High precision determination of atomic position is necessary for quantitative electron microscopy so that small width of peaks, which represent atoms in structural images, adequate resolution, and sufficiently strong image contrast are needed. The width of peak is usually determined by the point spread (PS) of instruments, but the PS of objects should also be taken into consideration in aberration-corrected transmission electron microscopy when point resolution of a microscope reaches the sub-angstrom scale, and thus the PS of the instrument is comparable with that of the object. In this article, PS is investigated by studying peak width with variation of atomic number, sample thickness, and spherical aberration coefficients in both negative Cs (NCSI) and positive Cs imaging (PCSI) modes by means of dynamical image simulation. Through comparing the peak width with various atomic number, thickness, and values of spherical aberration, NCSI mode is found to be superior to PCSI considering the smaller width.

Key words: point spread, peak width, aberration-corrected transmission electron microscopy, quantitative electron microscopy, negative Cs imaging, positive Cs imaging

INTRODUCTION

When aberrations in transmission electron microscope (TEM) objective lenses are corrected and resolution is sufficient to resolve atoms, precision becomes mandatory for quantitative high-resolution transmission electron microscope (HRTEM). According to the study of Van Aert et al. (2002), precision of atomic position obtained by means of HRTEM mainly depends on four factors: atomic interdistance, resolution of the microscope, dose of electron counts, and width of peaks, which represent atoms.

For a conventional TEM with point resolution of about 2 Å, peak width in optimum-focus images (structural images) is limited by the point spread (PS) of an instrument, the Fourier transformation of phase-contrast transfer function of the objective lens (Kirkland, 2010) according to the weak-phase object approximation (Scherzer, 1949). For an aberration-corrected transmission electron microscope (ACTEM) with point resolution increased to 0.5 Å, PS caused by an instrument is comparable to that caused by an object (Lentzen, 2008) so that the contribution of the latter cannot be ignored in determining peak width.

In this article, peak width in projected potential functions (PPFs), exit wave functions (EWFs), and optimum-condition images is determined, respectively, to study the PS in ACTEM. In TEM, the objective lens is below the sample so that the peak width in PPFs and exit planes can reflect the PS of objects, while the width in images should reflect the combination of the PS of both the object and instrument. The influencing factors as to peak width are studied, including atomic number, sample thickness, and spherical aberration coefficient in positive Cs imaging (PCSI) and negative Cs imaging (NCSI) modes (Jia et al., 2003, 2004; Jia & Urban, 2004). For the difficulty of studying the peak width analytically, the numerical method, image simulation comprising a full dynamical calculation of electron scattering, and contrast transfer under partially coherent illumination, are utilized. The full-width of the peak at half-maximum (FWHM) is determined by means of the Lorentz or Gaussian function fitting and is used to represent the peak width.

IMAGE SIMULATION

For simplicity, a fictitious model with face-centered cubic structure is used to investigate the peak width in this article. To avoid interaction between adjacent atoms, the lattice parameter of the model is set to \( a = 0.41 \) nm in the \(<001>\) direction, the projection direction simulated in this article, and the smallest interplanar distance set to 0.205 nm. Atomic species are varied to investigate the effect of scattering power, and a Debye–Waller factor of 0.005 nm\(^2\) applied, regardless of the atomic species in the simulation of the Multislice method (Cowley & Moodie, 1957). The main imaging parameters are an accelerating voltage of 300 kV, energy spread of 0.8 eV, semiconvergence angle of 0.1 mrad, and vibration of 0.03 nm in both \( x \) and \( y \) directions.

Moreover, the optimum imaging condition is corrected using an effective object plane close to the midplane of the sample instead of the exit plane (Bonhomme & Beorchia, 1983; Lentzen, 2008; Yu et al., 2011). That is, the focus value should be revised as \( \Delta f_{\text{corrected}} = \Delta f_{\text{sch}} - 0.5 t \) with \( \Delta f_{\text{sch}} \) the Scherzer focus and \( t \) the sample thickness. For example, for
Cs = −13 μm and wavelength 0.0197 Å, the Scherzer focus is calculated to be 5.8 nm, but when sample thickness, say 6 nm, is taken into consideration, the optimum-focus value used in this article for image simulation is 2.8 nm.

In addition, according to the theory of weak-phase object approximation, even an optimum-focus image may not reflect the true structure with increasing thickness. However, in reality, the peak position still accurately corresponds to the atom position if, according to the theory of pseudo weak-phase object approximation, thickness is below a certain value (Li & Tang, 1985). This is named as the critical thickness, meaning that atoms show no contrast for at that thickness. For example, in PCSI mode, atoms will show bright contrast above that thickness (Li & Tang, 1985). In this article, only the peak width is studied and only below the critical value so that optimum-focus images can still be treated as structure images unless noted otherwise.

Results

Peak Width in PPFs

Atomic species are selected randomly in their PPFs simulations and the variations of peak width in PPFs with different atomic numbers are shown in Figure 1. For most of the atomic species in our study, the peak width changes between 0.32 and 0.4 Å with the largest one about 0.48 Å. Figure 1 confirms the existence of object PS as proposed in Lentzen (2008), and it should be taken into consideration in determining the peak width in the image, especially when the resolution of the microscope is 0.5 Å, almost equal to the peak width in PPFs and even smaller than the peak width in EWFs with some thickness (to be introduced in the next subsection). From this standpoint, the ultimate image resolution obtained by an ACTEM may not be limited by the information limit of the instrument, but by the objects.

In addition, as shown in Figure 1, peak width oscillates with increasing atomic number, but with no obvious period or relationship between peak width and the atomic scattering power (Doyle & Turner, 1968). More work needs to be done to understand this phenomenon.

Peak Width in the EWFs

When electrons transmit through materials, especially crystals in the main axis, their trajectories change due to the electrostatic potential of the screened nucleus. Thus, the distance for electrons to transmit through the materials, i.e., the sample thickness, will influence the peak width in the exit plane, as described in the channeling effect (Van Dyck & Chen, 1999), where thickness influences the intensity of the peak.

Six atomic species, Li, C, Na, Si, K, and Ge with different scattering power, were selected at random, as marked in Figure 1, to study the peak width in the EWFs. Their width variations with different thickness are shown in Figure 2. The influence on PS by combination of atomic number and sample thickness in EWFs is found to be different from that by only atomic number in PPFs: the PS in EWFs is roughly dependent on the atomic number—atoms with larger atomic number correspond to a smaller peak width in EWFs. This feature is contrary to our perspective that heavy atoms usually have a large peak width and light atoms have a small width. In the following subsection it will be shown that this opinion is correct for the PCSI mode, but not for the NCSI one.

In addition, in Figure 2 it can also be observed that with an increase in sample thickness there is some fluctuation in peak width, which is similar to that described by the channeling effect (Van Dyck & Chen, 1999), where image contrast vibrates with thickness. However, no uniform period was found, even after detailed study with a smaller thickness interval or a much thicker sample.

Peak Width Variations in Images with Different Thicknesses

Electrons are reflected through the objective lens such that peak width in the image plane may be influenced by imaging
conditions such as spherical aberration. In this and the next subsections, influences on peak width by the sign and the absolute value of the spherical aberration, combined with sample thickness will be studied. C, Si, and Ge are used as examples.

A thickness series of images for Si are shown in Figures 3a and 3b, PCSI mode with $C_s = 0.013$ mm and NCSI mode with $C_s = -0.013$ mm, respectively. Thus, in Figure 3a, atoms display black contrast, while in Figure 3b they display white contrast. Obvious discrepancies can be observed between Figures 3a and 3b. First, the critical thickness in PCSI is much smaller than that in NCSI: about 6 nm in PCSI mode, as shown in Figure 3a, while for NCSI (Fig. 3b) it is almost five times larger than that in PCSI, about 32 nm. Second and more importantly, peak width in the PCSI mode increases with increasing sample thickness, while in the NCSI mode the peak width decreases at first with increasing thickness, which indicates the difference in PS in the PCSI and NCSI modes. Moreover, similar results can be obtained for C and Ge, which can be clearly seen in Figures 4a and 4b, which show the scatter plots of peak width versus thickness for PCSI and NCSI modes, respectively.

Moreover, in PCSI (Fig. 4a), the peak width of atoms with a large atomic number, such as Ge, increases more rapidly than ones with a smaller atomic number, such as C. In NCSI, this decreases more rapidly when the sample is not very thick (below 10 nm). That is to say, when the sample is thin enough, the larger the atomic number, the larger the PS in PCSI is, while in NCSI it is opposite.

Things become complicated for NCSI, however, when samples are thicker. Peak width fluctuates with increasing

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**Figure 3.** a, b: Thickness series of simulated images of Si with $C_s = 0.013$ and $-0.013$ mm, respectively.

**Figure 4.** a, b: Scatter plot of peak width for C, Si, and Ge in simulated image versus sample thickness with $C_s = 0.013$ and $-0.013$ mm, respectively. Bars represent the standard error.
thickness and its period seems to be connected to the atomic number. However, on the whole, the peak width in NCSI is always smaller than the width in PCSI for a certain species of atoms, at least in the scope of our research, which has been confirmed by ACTEM observation of the superconductor $\text{Bi}_2\text{Sr}_{1.6}\text{La}_{0.4}\text{CuO}_6+\delta$ (Ge et al., 2013). From this point of view, NCSI is superior to PCSI.

In addition, one feature should be noted in Figures 4 and 6: width value reaches almost 2 Å for PCSI and 1 Å for NCSI, which is much larger than the resolution obtained experimentally. This discrepancy may be caused by the definition of width, i.e., FWHM, but it is reasonable to use this definition if only the trend in peak width with respect to the various factors is discussed.

**Peak Width Variations in Images with Different Cs Values**

Figures 5a and 5b show a Cs series of simulated images of Si with sample thickness 2.4 nm in PCSI and NCSI modes, respectively. In general, the peak width decreases with decreasing absolute Cs values for both PCSI and NCSI modes, just as expected. More details can clearly be seen in Figure 6 that shows the scatter plots of peak width versus absolute Cs values with three thicknesses, 1.6, 2.4, and 3.3 nm. In Figure 6a, showing the PCSI mode, a rapid decrease in the peak width is found when Cs value decreases from 0.1 to 0.05 mm, but with further decrease no obvious changes occur. On the contrary, in NCSI mode (Fig. 6b), the peak width decreases gradually with decreasing absolute Cs value from 0.1 to 0.03 mm and then decreases steeply from 0.013 to 0.003 mm.

Moreover, similar results as from Figures 3 and 4 can be obtained when the sample is thin enough that peak width increases in PCSI with increasing sample thickness, while in NCSI it decreases, and moreover, peak width in NCSI mode is always smaller than that in PCSI mode for a certain atomic species.

**DISCUSSION**

Comparing with conventional TEM, the PS of ACTEM is complicated: NCSI and PCSI show a different trend with variation of thickness and atomic number. With an increase in thickness, PS in NCSI decreases when the sample is not too thick, but it is opposite in the case of PCSI. Analogically, PS in NCSI decreases with an increase in atomic number, while it increases in PCSI. These differences may be ascribed to the different contribution of the nonlinear imaging in NCSI and PCSI.

It is interesting that there is no obvious relation between atomic number and the PS in PPFs, but in EWFs the PS is dependent on the atomic number: PS decreases with an increase in atomic number. It indicates that it is the atomic columns in crystal materials that play a significant role in determining the PS, while the nucleus itself, or the scattering power, is not related directly to the PS.

Moreover, NCSI is a newly developed imaging mode in ACTEM (Jia et al., 2003, 2004; Jia & Urban, 2004) and has been proven to be superior to PCSI. Besides the well-known characteristics, such as enhanced image contrast (Jia et al., 2003, 2004) and signal-to-noise ratio (Jia et al., 2010), the smaller PS and standard error are also meaningful, as shown in Figures 4 and 6. Especially, when the sample is not too thick, the PS decreases in NCSI mode with an increase in sample thickness, while it always increases in PCSI.
Smaller PS and standard error, as well as the enhanced signal-to-noise ratio (Jia et al., 2010), can greatly improve the measured precision of the atomic position. In addition, another advantage of NCSI is the larger critical thickness as shown in Figure 3. The image taken with a thicker sample can be directly interpreted in NCSI, which is especially beneficial for observation of light atoms and weak structural signals.

**SUMMARY**

Described by peak width, PS in ACTEM has been investigated in PPFs, EWFs, and images, respectively. The following conclusions have been reached:

1. The PS of an object is confirmed by the peak width in PPFs. No obvious relationship is found in PPFs between PS and atomic number as well as the scattering power, but in the exit plane, a smaller PS has been generally found for heavier atoms, which means that not atomic number but atomic columns in the crystal materials play a significant role in determining the PS.

2. With an increase in sample thickness, PS decreases in the NCSI mode when samples are not too thick, but it always increases for PCSI. Compared with that of light atoms, the PS of heavy atoms decreases in NCSI mode and increases in PCSI more rapidly.

3. Generally, with decrease of the Cs value the PS decreases in both NCSI and PCSI modes, and on the whole, PS in NCSI mode is always smaller than that in PCSI mode for the same species of atoms.

Considering the smaller PS as well as the enhanced signal-to-noise ratio, NCSI is preferred for quantitative structural determination.

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