A practical approach for STEM image simulation based on the FFT multislice method

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Abstract

It has been demonstrated that a high-angle annular dark-field (HAADF) STEM technique gives an image resolving atomic columns. Due to the diffusion of this technique and an improvement of its resolution, a practical procedure for image simulation becomes important for a quantitative interpretation of the HAADF image. In this report a new practical scheme for a STEM image simulation is developed based on the FFT multislice algorithm. Here, a HAADF intensity due to thermal diffuse scattering (TDS) is calculated from the absorptive potential corresponding to high-angle TDS and the wave function equivalent to the propagating probe within the sample. Contrary to the commonly used Bloch wave method, a coherent bright-field intensity and a coherent HAADF intensity are also obtained straightforwardly. The HAADF image contrast calculated for GaAs is not simply proportional to $Z^2$ as expected from the Rutherford scattering at high-angle, and the As/Ga contrast ratio depends on the specimen thickness. This suggests that the generation of the HAADF signal is appreciably affected by the coherent dynamical scattering. The developed procedure here will have a definitive advantage over the Bloch wave approach for simulating the HAADF images expected from a defect and interface or amorphous materials, and also the HAADF image obtained by using a Cs-corrected microscope. This is because the former requires a huge super cell, while the latter needs a large objective aperture including a large number of incident beam directions. © 2002 Elsevier Science B.V. All rights reserved.

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1. Introduction

Crew and coworkers [1,2] have developed a scanning transmission electron microscope (STEM) and succeeded to detect an atomic cluster and even a single atom. However, the STEM has been used mainly for an analytical investigation at medium resolution. Recently, it has been demonstrated on a dedicated STEM that a high-angle annular dark-field (HAADF) technique [3] is capable of taking an image that resolves atomic columns [4,5]. This technique will become more important, because it has been shown that a conventional TEM with a STEM attachment can also give such a high-resolution HAADF image [6]. The importance of this technique will be further amplified by a recent development of a spherical aberration corrector for a probe forming system [7].
The formation of HAADF image has been explained as an incoherent imaging, which means that there is no interference between the scattered waves from different atoms or atomic columns. Since the major signal source of the HAADF image is a thermal diffuse scattering (TDS), the HAADF image is relatively immune to a defocus and/or thickness effect contrary to an image obtained with a conventional transmission electron microscope (CTEM) [8]. However, it has been also argued that even coherently scattered electrons will give an incoherent image [9,10,11]. The theoretical interpretation of HAADF images is thus still a subject of controversy. Recently, an unbelievable information transfer better than 0.078 nm has been claimed by using a microscope with a conventional point resolution of 1.9 nm [12]. On the other hand, it has been demonstrated that even a HAADF image gives significant artifacts by using image processing and image simulation [13]. Hence, an image simulation is as much important for evaluating an atomic resolution STEM image as for evaluating a high-resolution CTEM image.

However, STEM image simulation requires an enormous amount of computing power compared with CTEM image simulation, since we have to evaluate the dynamical scattering at each scanning point. Kirkland et al. [14] performed the first STEM image simulation in the late 1980s on a super computer taking into account only elastic scattering by using the multislice algorithm [15,16]. However, contrary to high-resolution CTEM image simulation, it is mandatory to consider the TDS that mainly determines the HAADF image contrast. Nevertheless, it is almost impractical to treat the TDS without any approximations, since the TDS is a time-dependent incoherent process. Therefore, several approaches to approximately treat the TDS have been proposed. These days the FFT multislice algorithm [16] is commonly used for simulating a high-resolution CTEM image. Contrary to this, so far the Bloch wave technique has been successfully applied for the HAADF STEM image simulation. This is because a series of approximations applied by Pennycook and Jesson [8] to the Bloch wave approach greatly reduces a computing burden. Using these approximations, the experimental images taken at a resolution just resolving atomic columns have been well explained. However, these approximations may not be applicable for an ultra high-resolution image to be taken with a Cs-corrected objective. Therefore, it is desirable to establish a new practical technique to include a TDS contribution within the framework of the multislice method. In this report, at first we will survey the approaches so far proposed to simulate a HAADF STEM image including the TDS contribution. Next, we will introduce a practical scheme for a STEM image simulation based on the FFT multislice algorithm. Using the program developed on the new scheme, then we will present some results calculated on the latest personal computer.

2. Survey on the procedures for HAADF image simulation

Some attempts have been made to include contributions from inelastic scattering into the STEM image simulation. We will focus on the TDS among inelastic events, since it is the most important inelastic contribution to the HAADF STEM image. Wang and Cowley [17] have generalized the multislice theory to include the TDS based on a set of coupled Schrödinger equations. Although some simplified approaches have been proposed later [18], there seems to be no other reports based on this treatment. Rose and coworkers [19,20] have developed a new slice algorithm by introducing a concept of mutual dynamic object transparency. Here, the thermal vibration is taken into account by averaging the mutual dynamic object transparency. On the other hand, the effect of thermal vibration has been emulated by using the atomic model randomly displaced from its averaged position, which is a snapshot of the thermally vibrating object [21,22]. In these schemes, thermally scattered electrons propagate through an object with elastic electrons, since an energy transfer to a phonon is negligible. Then, averaging many calculations with randomly assigned phases or displaced atoms is necessary to destroy an artificial interference between an elastic wave and a thermally scattered inelastic wave.
These approaches, therefore, require a substantial computing power.

Contrary to the above-mentioned approaches, Pennycook and Jesson [8] have proposed a more practical approach based on an absorptive potential [23]. Here, an integrated intensity falling on the HAADF detector is simply given by a sum of the probe intensity at each atomic column weighted by the corresponding TDS cross section. The limitations of their approximations will be discussed later compared with our new approach, which is a counterpart of this approach implemented by using the FFT multislice algorithm.

Nakamura et al. [24] have also used an absorptive potential to evaluate a decrease of the total intensity by using the multislice technique. Then, the decreased intensity is scattered over the detector plane by assuming the same Gaussian distribution function for all the scattering points. Another scheme called the ‘β-approximation’ has been implemented into a multislice procedure [25]. In this scheme, a coefficient β is determined by using the approach described by Dinges et al. [19]. Then, it is assumed that this coefficient is constant over the whole unit cell. Although these two approaches may be applicable to a specimen composed of a single element or elements with similar atomic numbers like GaAs, a different distribution function or β-coefficient may be required for a different scanning position when the difference of atomic numbers of constituent atoms becomes significant.

Amali and Rez [26] have recently developed a procedure based on their first-order perturbation theoretical analysis. This procedure seems to be promising, since it does not depend on the random process. However, the calculation time scales as the fourth power of the number of Bragg reflections included for dynamical calculation, since the dynamical elastic scattering is evaluated by using the Bloch wave method [26].

3. New practical approach for HAADF simulation

3.1 Procedure to estimate the TDS contribution

Our scheme for HAADF simulation follows the same line developed by Pennycook and Jesson [8] for a Bloch wave approach, but is adapted for the FFT multislice algorithm [16]. Here, we will briefly explain our scheme in the multislice framework, where a specimen is divided into a series of slices, each of which interacts with an incident electron successively [15].

The effect of the TDS may be treated by adding the imaginary component \( V''(r) \), an absorptive potential, to the normal electrostatic potential \( V'(r) \), which is now the real component of a total potential \( V(r) \)

\[
V(r) = V'(r) + iV''(r).
\]  

According to the multislice approximation, an incident wave \( \psi_{\text{in}} \) will be modified as follows by passing a thin slice:

\[
\psi_{\text{out}}(b) = \{\psi_{\text{in}}(b)q(b)\} \otimes p(b),
\]

where the vector \( b \) indicates a two-dimensional position over the slice, and \( \otimes \) designates a convolution integral. Here, \( p(b) \) is the propagator and \( q(b) \) the phase grating defined by

\[
q(b) = \exp\left\{ -\frac{i}{\hbar \nu} V_p(b) \right\} \\
= \exp\left\{ -\frac{i}{\hbar \nu} V'(b) \right\} \exp\left\{ \frac{1}{\hbar \nu} V''(b) \right\},
\]

where \( \nu \) is the electron velocity, \( \hbar \) the Plank constant divided by \( 2\pi \). \( V_p(b) \) is the complex potential within the slice projected along the incident beam direction, and \( V'_p(b) \) and \( V''_p(b) \) are its real and imaginary components, respectively. Since the magnitude of the propagator is unity for a normal incidence [27], we have the following relation for the magnitude of the Fourier transform of the output wave function \( \Psi_{\text{out}}(u) \):

\[
|\Psi_{\text{out}}(u)| = \left| FT\{\psi_{\text{in}}(b)q(b)\} \cdot P(u) \right| = \left| FT\{\psi_{\text{in}}(b)q(b)\} \right|.
\]

where \( u \) denotes the reciprocal space coordinate conjugate to \( b \). By using this relation and Parseval’s theorem [28] twice, we have the
following equality:

\[
\int |\psi_{\text{out}}(b)|^2 \, \text{d}b = \int |\Psi_{\text{out}}(u)|^2 \, \text{d}u \\
= \int |FT\{\psi_{\text{in}}(b)q(b)\}|^2 \, \text{d}u \\
= \int |\psi_{\text{in}}(b)q(b)|^2 \, \text{d}b.
\] (5)

The loss of electrons by passing the slice will then be given using this equality and Eq. (3) as

\[
I = \int \left( |\psi_{\text{out}}(b)|^2 - |\psi_{\text{in}}(b)|^2 \right) \, \text{d}b \\
= \int \left| \psi_{\text{in}}(b) \right|^2 \left[ \exp \left\{ \frac{2}{\hbar V^0_p(b)} \right\} - 1 \right] \, \text{d}b \\
\approx \int \frac{2}{\hbar V^0_p(b)} \left| \psi_{\text{in}}(b) \right|^2 V^0_p(b) \, \text{d}b.
\] (6)

This equation does not give any information about the distribution of inelastic electrons on the STEM detector. However, we are interested only in the integrated intensity of inelastic electrons over the detector. The TDS electrons falling on the HAADF detector can thus be easily calculated, when we use an absorptive potential corresponding to the high-angle TDS scattering (see the next section).

It is noted that the last expression of (6) is the fundamental used also by Pennycook and Jesson [8], who applied a series of approximation to replace the absorptive potential by the delta function. Our approach without these approximations will become indispensable for a very fine probe comparable with the width of the absorptive potential (see later Discussions). We neglect a further scattering of a thermally scattered electron, which may redistribute the TDS electrons. However, this single scattering approximation will not give an appreciable error, since we are not interested in a detailed scattering distribution itself, but an integration of the distribution. Even a strong Kikuchi line will not affect our image, if the detector covers a large angular range compared with the width of the Kikuchi line [8].

3.2. Complex potential and high-angle absorptive potential

The scattering potential is calculated from an inverse Fourier transform of the Fourier components \( V_\text{g} \) expressed by the atomic form factors \( f_k \):

\[
V_k = \frac{2\pi\hbar^2}{m_0} \Omega \sum_k f_k(s, M_k) \exp(-M_k s^2) \exp(-2\pi\imath r_k),
\] (7)

where \( f_k \) describes the stationary position of atom \( k \), \( m_0 \) the electron rest mass and \( \Omega \) the unit cell volume. The form factor is a function of \( s = \sin \theta/\lambda \), where \( \theta \) is the Bragg angle and \( \lambda \) the electron wave length, and \( s = g/2 \). Here, the Debye–Waller factor of the form, \( \exp(-M_k s^2) \), is extracted from the atomic scattering factor. Let us define an effective form factor, which is obtained by multiplying the normal form factor by the Debye–Waller factor. Then, actual scattering strength is controlled by the effective form factor.

The normal real potential \( V^0_p(b) \) will be evaluated by using the elastic atomic scattering factors (elastic form factors) tabulated in [29] or by using analytical expressions given by Doyle and Turner [30]. Most of HREM image simulation programs use analytical expression for the electron scattering factors derived from the Mott formula using X-ray scattering factors.

The total absorptive potential \( V^0_p(b) \) will be evaluated by using the following absorptive form factor \( f^0_k \) [23]:

\[
f^0_k(s, M_k) = \frac{4\pi\hbar}{m_0\hbar} \int f_k(\mid s')\mid f_k(\mid s - s') \mid \\
\times \left[ 1 - \exp\{-2M_k(s^2 - s \cdot s')\} \right] \, \text{d}^2s',
\] (8)

where \( f_k \) corresponds to the elastic form factor of atom \( k \). The integration in Eq. (8) should be taken out to an \( s \) value of about 30 Å\(^{-1}\) to achieve an accuracy of better than 99% [31]. However, the analytical expressions given by Doyle and Turner [30] are only valid up to \( s = 2.0 \) Å\(^{-1}\). Weickenmeier and Kohl [32] have developed another form of analytical expressions for elastic form factors, and also derived analytical formula for the total absorptive form factor based on Eq. (8).

The absorptive potential \( V^0_p(b) \) corresponding to high-angle TDS scattering will be obtained by
using the following absorptive form factor:
\[
f_k^{HA}(s, M_k) = \frac{4\pi \hbar}{m_0 v} \int_{\text{detector}} f_k(|s'|) f_k(|s - s'|) \left[ 1 - \exp\{-2M_k(s'^2 - s \cdot s')\} \right] \, d^2 s',
\]
where the integration is carried out only over the detector area [8]. This treatment is valid when an illumination is parallel or an incident cone angle is negligible. However, this treatment may be applied even for a rather large objective aperture required for a Cs-corrected STEM. This is qualitatively explained as follows by using Fig. 1, which illustrates the geometry of wave vectors and scatter vectors involved in the TDS scattering [23]. For the effective form factor obtained by combining the Debye–Waller factor and Eq. (9), the corresponding integrand will be rewritten as
\[
f_k \left( \frac{g' - t}{2} \right) f_k \left( \frac{g - g'}{2} \right) \left[ \exp\{-M_k (g - t)^2\} \right. \\
\left. \exp\{-M_k (g' - t)^2\} \exp\{-M_k (g - g')^2\} \right],
\]
where \( g - t \) and \( g' - t \) corresponds to the scattering vectors to \( G \) and \( G' \) in the case of the tilted incident beam direction. We may then note the followings. The underlined terms are constant assuming the fixed scattering directions \( k_g \) and \( k_{g'} \) for any incident directions. Since the form factor \( f_k \) is a monotonically decreasing function, its change owing to tilting a beam is partly compensated by the change of \( \exp\{-M_k (g' - t)^2\} \) in the square brackets. Moreover, in considerable cases
\[
\exp\{-M_k (\frac{g' - t}{2})^2\} \approx \exp\{-K_k s'^2\},
\]
because the inner edge of the high-angle annular detector is a few times larger than the size of the objective aperture even for a Cs-corrected situation [38]. For the scattering vector \( g \) in question, on the other hand,
\[
\exp\{-M_k (\frac{g - t}{2})^2\} \approx \exp\{-K_k s^2\}
\]
when it is large, while
\[
\exp\{-M_k (\frac{g - t}{2})^2\} \approx 1 \approx \exp\{-M_k s^2\}
\]
when it is small. Furthermore, for the scattering vector \( g \) the contributions to the form factor for a fixed \( g \) from a diametrical pair of incident beam directions \((+t \text{ and } -t)\) are apt to average out to the contribution from the central beam. In conclusion, the absorptive form factor for high-angle TDS scattering will be given by Eq. (9) within a reasonable approximation, even when an objective aperture is rather large.
3.3. Probe forming including wave aberrations

For the STEM image simulation by using the multislice technique the wave function of the electron probe $\psi_p(b; b_0)$, which will propagate down the specimen, is calculated at the entrance surface from the following Fourier transform

$$\psi_p(b; b_0) = \int_{\text{aperture}} \exp(-iW(k))\exp(-2\pi i k(b - b_0))dk,$$

(10)

where $b_0$ describes the probe position and $k$ the wave vector equal to $2\sin \theta/\lambda \approx x/\lambda$ for the scattering angle $x$. The integration is carried out over the objective aperture. Here, $W(k)$ is the wave aberration function of the objective (probe forming) lens

$$W(k) = \frac{2\pi}{\lambda} \left( \frac{1}{4} Cs(\lambda k)^4 - \frac{1}{2} Z(\lambda k)^2 \right),$$

(11)

where $Cs$ and $Z$ denote the third-order spherical aberration coefficient and the defocus of the objective lens, respectively.

3.4. Calculating STEM image signals

Three STEM signals, i.e., coherent bright- and dark-field intensities and HAADF TDS intensity are evaluated at successive specimen thickness for each scanning position. Here, we take the advantage of the FFT-multislice algorithm that can evaluate both the wave function $\psi_p(b)$ and its Fourier transform $\Psi_n(u)$ at each slice. The coherent bright- and dark-field intensities from the specimen of $n$ slices are simply calculated by using the scattering amplitude $\Psi_n(u)$ at the last slice from the following equations:

$$I_n^{BF} = \int_{BF} |\Psi_n(u)|^2 du,$$

(12)

and

$$I_n^{DF} = \int_{DF} |\Psi_n(u)|^2 du,$$

(13)

respectively, where the integrations are carried out over the bright-field detector or the dark-field detector. The HAADF TDS intensity from the specimen of $n$ slices is calculated by adding the TDS intensity generated from each slice down to the last slice. Here, the TDS signal is easily calculated from the wave function $\psi_j(b)$ and a high-angle TDS absorptive potential $V_j^{HA}(b)$ (see Eq. (6)) as follows:

$$I_n^{HA} = \sum_{j=1}^{n} \frac{2}{\hbar v} \int |\psi_j(b)|^2 V_j^{HA}(b) db,$$

(14)

where the integration is carried out over the whole sampling points in real space.

4. STEM image simulation

We choose here GaAs [011] as an example to demonstrate the feasibility of the procedure developed here. In all cases shown below we assume a 200 kV-microscope with $Cs = 0.5$ mm and an annular dark-field detector that collects electrons scattered between 56 and 100 mrad ($s = 1.12$ and 2.0 Å$^{-1}$). The Debye–Waller factors used for Ga and As, where otherwise stated, correspond to the displacement parameters 0.637 and 0.685 Å$^2$, respectively [33].

We calculate the real and imaginary potentials by using the elastic and total absorptive form factors of Weickenmeier and Kohl [32], respectively. The high-angle absorptive form factor is evaluated from the elastic form factor of Weickenmeier and Kohl [32] by using Eq. (9). These three form factors corresponding to the Gallium atom are illustrated in Fig. 2. They are all the effective form factors, that is, the form factors multiplied by the Debye–Waller factor. We may note again here that the effective form factor controls an actual scattering strength. The high-angle absorptive form factor shown here corresponds to the detector that collects electrons falling on between $s_1 = 1.0$ and $s_2 = 2.0$ Å$^{-1}$, which correspond to 50 and 100 mrad, respectively, for 200 kV electrons. It is shown in Fig. 2 that the effective high-angle absorptive form factor attenuates most slowly with the scattering angle. This figure indicates that all the three projected potentials for Ga will be evaluated with a sufficient accuracy when we include the Fourier components up to $s = 2.0$ Å$^{-1}$. 
Fig. 3 shows the elastic and total absorptive (real and imaginary) potentials, and the high-angle TDS absorptive potential calculated by including the Fourier components up to 4.0 Å⁻¹ \((s = 2.0 \text{ Å}^{-1})\). It is clear that the high-angle absorptive potential is significantly sharper than the elastic potentials but is substantially different from a point-like distribution.

Next, we determine the optimum objective apertures by using Rayleigh’s quarter wavelength rule [34], which says that the quality of the image is not seriously affected when the wave-front deformation is less than a quarter of a wavelength. Fig. 4 illustrates the frequently used two cases, namely, the Scherzer conditions [35] and the Gabor conditions [36]. The former conditions are attained by limiting the wave aberration between 0 and \(\frac{1}{4}\lambda\), resulting in an objective aperture of 11.9 mrad at \(Z = 354 \text{ Å}\). On the other hand, the later conditions are realized by an objective aperture of 11.0 mrad at \(Z = 250 \text{ Å}\), which limit the wave aberration between \(-\frac{1}{8}\lambda\) and \(\frac{1}{8}\lambda\). The latter conditions are close to the ones proposed by Mory et al. [37]. Fig. 5 shows the probe shapes corresponding to these two conditions. The first minimum in both cases happens to appear closely at the shortest distance between Ga and As on the dumbbell. The Scherzer conditions are superior to the Gabor conditions in terms of the peak height as well as the peak width. We use thus the Scherzer
conditions for the later simulations for GaAs. However, the Gabor conditions may be more appropriate for analytical studies such as electron energy loss spectroscopy (EELS), since they give the lower side-lobe intensity than the Scherzer conditions.

For the STEM image simulation of GaAs [011], we use a matrix of $512 \times 512$ to cover a super cell made from $12 \times 8$ [011] unit cells, which extends over 48.0 by 45.1 Å. A real space sampling interval then corresponds to about 0.1 Å, and the Fourier space extends up to almost $5.0 \, \text{Å}^{-1}$ with a sampling interval of about $0.02 \, \text{Å}^{-1}$. The number

![Wave aberration functions for the Scherzer conditions (solid line) and the Gabor conditions (broken line). Vertical bars with arrowheads on both sides indicate the ranges satisfying Rayleigh’s quarter wavelength rule. The probe shapes corresponding to these two conditions are shown in Fig. 5.](image1)

![Probe shapes corresponding to the Scherzer (a) and Gabor (b) conditions. Left half and right half of these probes are, respectively, shown here. Horizontal scale indicates 0.5 nm.](image2)

![HAADF-simulated images for GaAs [011]. We assume a 200kV microscope with $C_s = 0.5 \, \text{mm}$, an objective aperture of 11.9 mrad and an annular dark-field detector extending between 56 and 100 mrad. The specimen thicknesses for (a)–(c) correspond to 40, 120 and 200 Å, respectively. Each image is normalized between its intensity range: (a) 0.003–0.035, (b) 0.012–0.098 and (c) 0.023–0.129, where the highest intensity corresponds to white. Image appearance does not change so much except an overall growth of the signal strength as well as the background. The width of each figure corresponds to 5.64 Å.](image3)
of incident beam directions within the objective apertures is more than 1400. We may note that the scattering from all these incident beam directions can be simultaneously calculated by propagating the probe by using the multislice technique. A scanning interval is arbitrarily defined and a scanning scheme can be selected from the area, line and point modes. A computation time on a PC\(^1\) is about 1 min for each scanning point calculated up to 50 slices. The area scan then takes about 11 h, where the scanning area, which is reduced to one quarter of the unit cell by utilizing the image symmetry \(cm\), is sampled by 21 \(	imes\) 29 points with an interval of approximately 0.1 Å.

The HAADF simulated images are reproduced in Fig. 6. These images do not change so much their appearances up to 20-nm thickness shown here except an overall growth of the signal strength as well as the background. Note that each image here is normalized between its maximum and minimum intensities to utilize the full range of gray scale.

Since the STEM image simulation requires an enormous amount of computing power, the line scan mode is sometimes very useful. The change of contrast with specimen thickness can be continuously displayed by using the line scan data. We will call this type of display “the thickness map” hereafter. The coherent bright-field and high-angle dark-field images are shown in Fig. 7 as thickness maps down to 200 Å. Here, the scan is carried out on a line along a GaAs dumbbell in the middle of the model. The bright-field images are calculated by assuming the detector that extends the same range of the objective aperture (11.9 mrad). The appearances of both images change with thickness in contrast to the HAADF images shown in Fig. 6. The high-angle coherent signal fades out with thickness due to dynamical extinction as well as absorption. It demonstrates, however, that a bright-field image and a high-angle coherent image showing similar features to the high-angle TDS image appear at certain ranges of specimen thickness.

### 5. Discussions

#### 5.1. High-angle absorptive potential

Pennycook and Jesson [8] have shown that for an annular detector with a large inner angle the
integral of Eq. (9) becomes a constant as defined by

\[ f^\text{HA}_k(M_k) = \frac{4\pi^2}{m_0 v} \int_{\text{detector}} f_k(s')^2 \left[ 1 - \exp\left(-2M_k s'^2\right) \right] 2\pi s' \text{d}s', \]

and replaced the high-angle absorptive potential by a delta function. It is seen from Fig. 2 that the form factor \( f^\text{HA}(M) \) for Ga is actually almost constant up to the inner edge of the dark-field detector \((s_1 = 1.0 \text{ Å}^{-1})\), if we note that the effective form factor attenuates in the same way as the Debye–Waller factor. However, Fig. 3 clearly demonstrates that the high-angle TDS absorptive potential is not an ideal delta function, since the potential is determined by the effective form factor. Moreover, an attenuation of the form factor \( f^\text{HA} \) itself becomes appreciable for the scattering vector larger than the inner detector edge. This is because the integrand in Eq. (9) becomes negative inside a circle having the scattering vector as its diameter [8]. Nevertheless, the delta function approximation may be acceptable and explain an experimental image, when the scanning probe distribution varies slowly compared with the absorptive potential. In passing it may be pointed out that the integral of Eq. (15) gives an exact scattering cross section, only when the delta function approximation is valid.

The experimental HAADF images have been so far explained by approximating the TDS absorptive potential with a delta function e.g. [8,13]. The achievable resolution under this approximation should be solely determined by the probe width. However, this approximation may not be applicable for a fine scanning probe attained by correcting the third-order spherical aberration [38]. Recently, Pennycook, Rafferty and Nellist [39] have suggested that the HAADF image shows the 1s Bloch states for a very fine probe, and thus argued that the resolution is limited by the width of the 1s Bloch states. We claim, however, that the achievable resolution will be restricted by the widths of the high-angle TDS potential.

5.2. HAADF image contrast

The contrast of the HAADF image has been explained by Rutherford scattering at high angle that predicts the \( Z^2 \) dependence [8]. This dependency is quantitatively explained by Eq. (15), which approaches to the value proportional to \( Z^2 \) for an annular dark-field detector with a large inner angle. Amali and Rez [26] obtained the As/Ga contrast ratio close to the expected \( Z^2 \) dependence (1.13) by assuming an 80 mrad inner detector angle for 100 kV accelerating voltage. According to our results, however, the contrast ratio of As/Ga is 1.08–1.09 up to about 50 Å, and decreases with a specimen thickness as indicated by square points in Fig. 8. Thus, the contrast ratio of As/Ga is always smaller than the expected \( Z^2 \) ratio. This may be partly explained by the effective high-angle TDS form factors as illustrated in Fig. 9. The form factor ratio As/Ga at the zero scattering angle is 1.12, which is close to the expected \( Z^2 \) ratio. However, this ratio decreases with increasing the scattering vector, since the

![Fig. 8. Contrast ratio of As/Ga as a function of specimen thickness. The HAADF images are calculated for two sets of the displacement parameters. Squares and dots show the contrast ratios obtained for the theoretical estimates (0.637 and 0.685 Å² for Ga and As) and the measured displacement values (0.687 and 0.568 Å² for Ga and As), respectively. The expected As/Ga ratio due to the Rutherford scattering predicting \( Z^2 \) dependence is 1.13.](image)
displacement parameter used for As is larger than the one used for Ga.

According to Reid [33] the experimental thermal displacement parameter of As is smaller than the one of Ga (0.687 and 0.568 Å² for Ga and As). Therefore, we re-calculate the HAADF images with these experimental displacement parameters. The obtained contrast ratio in this case is shown by dots in Fig. 8. The HAADF contrast ratio is thus rather sensitive to the Debye–Waller factors. It may be expected that a larger thermal displacement parameter will result in a stronger thermal scattering and thus a higher HAADF contrast. However, it is not so straightforward. The effective TDS form factor for a large displacement parameter is large at small scattering angle, while it decreases rapidly as a function of s. Thus, the high-angle absorptive potential for a small displacement parameter becomes sharp and its peak height is apt to become high. The elastic potential also becomes sharp for a smaller displacement parameter, which concentrates more electrons into its center due to dynamical scattering. Dynamical scattering will then increase an efficiency of thermal scattering from an atom with a small thermal displacement. The dependency of the contrast ratio on the specimen thickness strongly suggests a significant effect of coherent dynamical scattering on the generation of the HAADF signal. This advises us against simply using the high-angle form factor $f_{\text{HA}}(M)$ for a quantitative analysis of the HAADF image intensity.

Nellist and Pennycook [11] have argued that even a pure coherent dynamical scattering yields an incoherent HAADF structure image of the atomic columns. Fig. 7 demonstrates that a high-angle coherent image similar to the high-angle TDS image appears at certain ranges of specimen thickness. However, these ranges are difficult to be predicted, because they are complicatedly affected by the dynamical scattering. Moreover, the image contrast is not so immune to the specimen thickness, and will not show a simple dependence on the atomic number as expected for the HAADF image.

5.3. Bloch wave method vs. Multislice method

The STEM image simulation procedure based on the multislice algorithm requires a relatively large super cell of about $50 \times 50 \text{ Å}^2$ that is sampled with $512 \times 512$ points. Since this is required to sample the real and reciprocal spaces with sufficient accuracy, we cannot reduce this requirement even when simulating the STEM image for a sample with a small unit cell. Nevertheless, when a structure model requires a larger super cell than $50 \times 50 \text{ Å}^2$, the calculation for each scanning point will be carried out by using a region of $512 \times 512$ points cut out from the whole matrix. Thus, a calculation time for each scanning point is not affected by the size of the structure model in contrast to the Bloch wave method.

Another significant potential of the multislice approach is that the scattering from all incident beam directions within the objective aperture can be simultaneously calculated. Using the Bloch wave method, the matrix equation should be solved for each incident beam direction. This feature may become of great importance, when we have to open up the objective aperture by correcting the spherical aberration. It may be noted that Pennycook, Rafferty and Nellist [39] have simulated the images for an aberration-corrected STEM not by solving the matrix equation for each beam direction but by convoluting the free-space probe with a 1s Bloch state.
The Bloch wave method can calculate a HAADF STEM image at any thickness after solving the matrix equations. The multislice approach, however, requires a slice-by-slice evaluation of the scattering up to the required thickness. Therefore, the Bloch wave approach may be suitable for estimating the images expected from rather thick specimen. By using the multislice approach, however, all information up to the calculated thickness is in our hand at the end of calculation.

Further advantage of the multislice method is that the coherent bright-field image and the coherent dark-field image can be obtained with a negligible amount of additional computation in addition to the HAADF TDS image. It may be noted that no coherent STEM image simulation has been reported so far by using the Bloch wave approach.

6. Conclusions

A new practical scheme for a STEM image simulation has been proposed based on the FFT multislice approach, where a wave function within a sample and its Fourier transform are evaluated simultaneously. An integration of the TDS signal falling onto the high-angle dark-field detector is then easily estimated from a wave function and a high-angle TDS absorptive potential. Contrary to the commonly used Bloch wave method, a coherent bright-field image and a coherent contribution to the HAADF image are also straightforwardly obtained from the Fourier transform of the wave function.

Our STEM image simulation procedure requires a relatively large super cell of about 50 × 50 Å sampled with 512 × 512 points. However, when a structure model requires a larger super cell than 50 × 50 Å², a region of 512 × 512 points cut out from the whole matrix will be used for the calculation for each scanning point. Thus, the FFT multislice approach developed here has a definitive advantage over the Bloch wave approach for a simulation of a defect, interface or amorphous material. Furthermore, our procedure will be of great importance for the simulation of a Cs-corrected microscope, since we can simultaneously calculate the scattering from all incident beam directions within the objective aperture.

HAADF images as well as bright-field images are calculated for GaAs [011] by using the program developed on the new scheme, assuming a 200 kV-microscope with Cs = 0.5 mm. The image simulation for GaAs demonstrates that the HAADF image contrast may not be simply proportional to Z² as expected from the Rutherford scattering at high angle. This is partly explained by the fact that the Debye–Waller factor affects the effective form factor for the high-angle TDS in opposite ways at small and large scattering regions. The contrast ratio that depends on the specimen thickness strongly suggests that coherent dynamical scattering play an important role for the generation of the HAADF signal.

The proposed scheme here for a HAADF STEM image simulation is not a rigorous approach but is based on the approximation for the high-angle TDS form factor that is also assumed in other approaches. However, our approach will be applicable in a practical case, since high-angle TDS form factor changes rather slowly with a scattering angle. Since the developed procedure here has a definitive advantage over the Bloch wave approach, it will be used to resolve some controversial issues regarding the STEM image formation.

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