Full-length paper

New scheme for calculation of annular dark-field STEM image including both elastically diffracted and TDS waves

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Abstract
A new scheme of calculation of high-angle annular dark-field STEM image, capable of including both elastically diffracted and thermal diffuse scattering waves, has been presented by a combination of Pennycook’s and Nakamura’s methods. The new scheme has been demonstrated for image simulations of Si(011) as functions of thickness, defocus values and detector angles. In the present method, the TDS electron intensities are treated in the same way as in Pennycook’s method, having a clear physical picture of its origin and reflecting the atom configuration in the systems. For the case of Si(011), it has been confirmed that at the detector angle of 60 to 160 mrad, which is usually applied, the image becomes highly incoherent, and even the image formed only from SOLZ beams becomes incoherent at the detector angle. At a low detector angle, however, the image has coherent features indicating the necessity of a simulation for individual systems.

Keywords
scanning transmission electron microscope, Bethe method, multislice method

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Introduction
Since the image of isolated heavy atoms was obtained using a scanning transmission electron microscope (STEM) equipped with an annular detector [1], the method has been developed continually, and electron probes of atomic dimensions have become available [2–4]. By using a high-angle, wide range annular detector, the image is mainly formed from thermal diffuse scattering (TDS) electrons, and the coherent contribution of the image is averaged to be an incoherent image with a strong Z contrast. Therefore, images obtained from annular dark field (ADF)-STEM are expected to be intuitively interpreted with no contrast reversals along with the defocus and/or specimen thickness changes, in contrast with the phase contrast images in HRTEM [5]. However, the image made from coherent electrons still shows the coherent nature, and strongly excited diffracted beams and/or channelling of probe electrons may play an important role in the image in some situations. These conditions in various cases have been studied extensively by many authors [6,7]. Therefore, the appropriate selection of parameters, such as defocus and detector angles, is essential in ADF-STEM observations. It means that the image simulation of ADF-STEM image is still needed in order to achieve the incoherence of the image. Recently, many attempts to determine the density and the location of an impurity from a slight intensity difference of ADF-STEM image have been performed [8–10], so that a demand for the accurate and fast simulation method is increasing, and those calculation methods are studied by several authors [11–15].

Pennycook and Jesson [11] presented a scheme that evaluates the intensities of TDS electrons by summing up a single-atom-inelastic-scattering-factor obtained by Einstein model among the detector direction. The summed scattering factor is then multiplied by the amplitude of wave function at each atom site excited by the focused probe, calculated by the Bethe method. In the method, multiple scattering after TDS is ignored but it is justified for usual specimen thickness. Although the origin of the TDS waves amplitude is clear in this method, only the intensity relations between each probe position can be obtained because only amplitude at atom site is multiplied to the scattering factor. Therefore, it is impossible to take into account the elastically diffracted waves.

By using the multislice method [16,17], Loane et al. [13] discussed the visibility of a single ad-atom on semiconductor surface using a frozen phonon method. By this method, small deviations of each atom from its atomic site were introduced to express thermal vibration of atoms, and multislice calcula-
tion was performed for the configuration. TDS electron intensities are obtained by ensemble averaging of calculated intensities for many different configurations of atoms. This method also has a very simple physical picture of origin of TDS intensity, although the time for the calculation is rather long. Recently, Nakamura et al. [15] presented a new method based on the multislice method. In this method, the total absorbed amplitude of focused probe is first calculated and then the absorbed intensity is multiplied by the Gaussian distribution function which expresses the distribution of inelastically scattered waves under Fokker-Planck approximation.

For the electron wave function inside a crystal, both the Bethe and multislice methods give the same result for the same system [18]. For HRTEM image simulation, the multislice method usually has an advantage in its computing time. For the ADF-STEM image simulation, however, Bethe’s method is much faster than the multislice method for a small unit cell system. In the case of ADF-STEM image simulation by the multislice, one needs to repeat the whole calculation at each scan point of the probe. In the case of Bethe’s method, on the other hand, once the eigenvalues and eigenvectors have been obtained for each incident angle of probe, wave functions at each scan point can be readily obtained. Recently, Nellist and Pennycook [12] improved the method for the case of elastically diffracted electron intensity by Fourier transforming the wave function with a spatial frequency vector, and the efficiency of the calculation by the method is much increased. The method, however, is not capable of TDS electron intensities.

In this paper, a new scheme for calculation is presented by a combination of Pennycook’s and Nakamura’s methods. One of the unique advantages of this method is the fact that the elastically diffracted beam intensities can be evaluated together with TDS intensities. The TDS intensities are treated by the same method as that proposed by Pennycook’s group, having a clear physical picture of its origin. This method can be applied essentially in both the multislice and Bethe methods. The feature and effect of elastically diffracted waves in ADF-STEM image are discussed as functions of detector angle and defocus values for Si having [011] zone axis.

Theory

Electron wave function inside the crystal by focused probe

The electron wave function inside the crystal excited by a focused probe is the same as that obtained by the calculation of convergent beam electron diffraction (CBED). Here, we use notation similar to that used by Pennycook and Jesson [11]. Real and reciprocal space vectors are written in their components perpendicular and parallel to the optic axis, thus \( r = (R, z), \) \( k = (K_p, k_z) \). The wave function at position \( R_0 \) can be described as the summation of the Bloch waves excited by focused probe at position \( R_0 \),

\[
\psi (R - R_0, z) = \sum_{\alpha} A_\alpha (R - R_0, z) \tag{1}
\]

where \( A_\alpha \) denotes the Bloch wave of \( \alpha \)-th branch and can be written explicitly as

\[
A_\alpha (R - R_0, z) = \int_{\text{proj}} \phi_\alpha (R, z, K) \exp \{ i ( - K_\alpha \cdot (R - R_0) + W(K)) \} dK \tag{2}
\]

where \( W(K) \) expresses the phase factor due to the aberration of probe forming lens and the crystallographic sign convention is used in this paper. The integration is performed within the range of semi-angle of probe forming lens.

TDS intensity by Pennycook’s method

Pennycook and Jesson [11] showed TDS intensity at probe position \( R_0 \) can be obtained as a function of specimen thickness \( t \) as

\[
I_{\text{TDS}}^{\text{film}} (R_0) = \sum_{\alpha} \rho_{\alpha} | \psi (R_0 - R_0, z) |^2 dz \tag{3}
\]

using a delta-function approximation where \( \rho_\alpha \) represents the high-angle cross-section of atom X at \( R_0 \) positions. This can be expressed as follows using the Einstein model

\[
\rho_\alpha = \left[ 4\pi (m/m_0)/(2\pi\lambda) \right] \left[ \text{factor} | f_\alpha (s) |^2 | 1 - \exp(-2M_\alpha (s)) | ds \right] \tag{4}
\]

where \( f_\alpha (s) \) represents the scattering factor of X atom and \( M_\alpha (s) \) is Debye factor. The integration is performed among the detector range. In the case where an atom column contains different species of atoms, the summation of atom site can be expanded also in \( z \) direction by introducing \( z_r \). Under this \( \delta \) function approximation, the amplitude of wave function only at each atom site is used to obtain the TDS electron intensity from the atom. Thus, the integration of whole space is reduced to the summation of each atom site. Although this is a quite reasonable assumption, only the relative intensity for each scan point can be obtained.

TDS intensity by Nakamura’s method

In Nakamura’s method, the TDS intensity is obtained by a different concept from that in Pennycook’s method. The decay of the intensity of incident wave by absorption is usually expressed by introducing an imaginary part into a crystal potential. The non-Hermitian matrix produces the imaginary part of the eigenvalue and causes the damping of intensity. The total absorbed intensity \( I_{\text{abs}} \) of incident wave by crystal is first obtained by comparing the intensities of incident waves and transmitted waves. This \( I_{\text{abs}} \) expresses the magnitude of inelastic scattering of incident waves, therefore, some fraction of this \( I_{\text{abs}} \) will reach the detector. Nakamura et al. calculate this fraction by assuming the Gaussian distribution function \( g_{BG} \) of inelastically scattered waves as
\[ g_{BG}(s, t) = \frac{1}{\pi t^2} \exp(-s^2/2r(t)) \]

\[ \delta(t) = \sum_{j} \rho_j \left( \int_{0}^{t} \int_{0}^{r} f_j(s) ds \right) \]

where \( t \) is the total specimen thickness and \( \rho_j \) is the atomic composition of each \( j \) type element in the whole specimen and \( f_j(s) \) is the absorptive form factor of \( j \) atom element. The fraction of the intensity within the detector can be obtained by integrating this \( g_{BG} \) among detector direction as

\[ g_{HA} = \frac{1}{\text{Detector}} \int_{0}^{t} g_{BG} ds \]

By multiplying this \( g_{HA} \) to \( I_{abs} \), the intensity distribution of inelastically diffracted wave can be obtained.

New scheme of calculation

In a new method, instead of \( g_{HA} \), the ratio of TDS intensity within the detector to total TDS intensity is defined as

\[ g = \frac{\sum_{i} \sigma_{x}^{T} \int_{0}^{t} \left| \psi(R_i - R_{0}, z) \right|^2 dz}{\sum_{i} \sigma_{x}^{T} \int_{0}^{t} \left| \psi(R_i - R_{0}, z) \right|^2 dz} \]

where \( \sigma_{x}^{T} \) is the atomic cross section of TDS electron intensity for \( X \) atom species. For a plane incident wave scattering, this can be obtained by integrating the diffuse scattering distribution from an isolated atom

\[ \sigma_{x}^{T} = \left[ 4\pi(m/m_{0})/(2\pi\lambda_{0}) \right] \int_{0}^{t} \left| f_j(s) \right|^2 \left[ 1 - \exp(-2M_j(s)) \right] ds \]

In eq. 8, the \( \delta \) function approximation is still used, therefore the amplitude of wave function only at atom site is used to evaluate this \( g \) value. However, the distribution function \( g \) is now in the form of ratio of HAADF intensity to total TDS electron. Therefore, the TDS intensity is expressed by multiplying this \( g \) value to the total absorbed intensity \( I_{abs} \). Because this obtained TDS intensity accords with the elastic diffracted beams, the total intensity can be obtained as

\[ I_{HA}^{Total} = I_{Elastic}^{HA} + g \times I_{abs} \]

where \( I_{Elastic}^{HA} \) expresses the total intensity of elastically diffracted wave within the detector. \( I_{abs} \) needs to be calculated using the imaginary potential including only TDS scattering because the distribution \( g \) is constructed only from TDS distribution, otherwise, the calculation becomes inconsistent. The inelastic potential including only TDS becomes consistent with Nakamura’s method. In other words, when the system consists of more than one atom species, the \( g \) has a dependence on atom arrangements. This effect is not included in Nakamura’s Gaussian distribution.

Simulation conditions

Bethe’s eigenvalue method modified by Nellist and Pennycook [12], was used for the calculation in order to obtain both absorbed intensity and elastically diffracted wave intensities. Calculations with 193-beams of excitation error \( s_e < 25 \ [1 / \text{nm}] \), including second Laue zone (SOLOZ), was performed for each of 512 incident partial plane waves within an illuminating cone of semi angle 12 mrad. The acceleration voltage was 200 kV. With this semi angle setting, the radius of the disk was about 4.8 \ [1 / \text{nm}] \ while the half of the distance of (111) spots was about 1.6 \ [1 / \text{nm}] \, so the disks fully overlapped each other. The spherical aberration \( C_s \) used was 1.0 mm. Weickenmeier’s absorptive scattering factor [19] was used and the Debye-Waller factor for Si was estimated using thermal diffuse scattering data by Radi [20] as 0.00355 \ [\text{nm}^2] \. The detector angles of 20 to 53 mrad and 60 to 160 mrad were used to demonstrate the effect of elastic waves in an ADF image.

Results and discussion

Figure 1 shows the image of Si(011) formed only from absorbed intensities for three different thicknesses of 20, 50 and 80 nm. For the system consisting of only one atom species, an ADF-STEM image by TDS electrons is expressed by an absorbed intensity image multiplied by a constant value decided by detector angle. Thus, the image expresses an image made by TDS electrons besides its total amplitude. The brightness of the images was adjusted for each thickness to show the detail of the images. The value of the intensification is shown right side of the images, such as ‘× 4’ represents that the image intensified by 4 times. Therefore, the images at 80 nm thickness were 4 times brighter than at 20 nm. This result shows that the absorption increases with increasing thickness. When an ADF-STEM image is incoherent, the image can be understood by the simple convolution of probe function and the projected potential. The images in Fig. 1 have a focus
dependence, such as the Si dimmer seen as one elongate spot at defocuses 40 and 60 nm becomes 2 peaks at 80 nm; however, this can be understood by the shape change of the probe function. In all images of three focuses, the atom positions always appeared bright. Moreover, regardless of the total intensity, the image has almost no thickness dependence. These features of the image indicate that the image is incoherent. Weak false spots between Si columns are seen in the image at defocus 80 nm. This may be explained by the effect of the subsidiary peaks of the probe as pointed out by Watanabe et al. [21].

In Fig. 2, the images formed from elastically diffracted electrons with a low detector angle of 20 to 53 mrad are shown. The images and their thickness and defocus dependence are quite different from Fig. 1. As the defocus value changes, so does the image, and bright areas do not always correspond to the atom positions. The image also has strong thickness dependence. It should be noted that the image in the thickness of 50 nm is the brightest one of the three thicknesses. This shows that the effect of elastically diffracted waves also has complex thickness dependence differing from the case of TDS image. Therefore, the contribution of elastic waves toward total ADF-STEM image may also have complex thickness dependence and cannot be intuitively interpreted.

Figure 3 shows the image of elastically diffracted electrons with high angle detector of 60 to 160 mrad. At this detector setting, the SOLZ ring is inside the detector, but still the image becomes incoherent by the effect of the high angle detector.
ting, the SOLZ ring is within the detector and, actually, almost all the intensity is coming form the SOLZ ring, and still the image becomes incoherent by the filtering effect of a high angle detector. A detector setting of 60 to 160 mrad is usually used for Si observations. This result confirms that at this detector angle, both images formed form TDS and elastically diffracted electrons will become completely incoherent.

Total ADF-STEM images, including both TDS and elastically diffracted electrons, are shown in Fig. 4 with (a) low detector angle and (b) high angle. At the low angle detector setting, the intensity of elastic waves is larger, about 3 times, than TDS one so that the resultant images are similar to those in Fig. 2. For the high detector angle, because the intensities of elastic waves are so weak compared to the TDS ones, image is almost exactly same as TDS one.

**Concluding remarks**

A new scheme of calculation combining Pennycook’s and Nakamura’s methods is presented. By using this method, the effects of elastically diffracted beams can be evaluated together with TDS intensities. The method was demonstrated for image simulations of Si(011) as a function of thickness, defocus values and detector angles. In this method, the TDS intensities are treated by the same way as by Pennycook’s group, having clear physical picture of its origin and is able to accommodate the atom configuration of systems which could not be treated in Nakamura’s method. This method can essentially be applied in both the multislice method and the Bethe method, therefore, one can choose both of these methods depending on the system to be calculated. For the case of Si(011), detector angle of 60 to 160 mrad, which is usually applied, was highly incoherent, and even SOLZ beams also formed an incoherent image at the detector angle. At low detector angle, however, the image formed mainly by elastically diffracted waves and had strong focus and thickness dependence. The effects of the elastic waves also depended on a thickness, thus, it is necessary to perform the simulations to ensure the observing condition is adequately incoherent for each system.

**References**

a Si crystal by atomic-resolution high-angle annular dark field STEM. 


