An efficient way of including thermal diffuse scattering in simulation of scanning transmission electron microscopic images

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Abstract

We propose an improved image simulation procedure for atomic-resolution annular dark-field scanning transmission electron microscopy (STEM) based on the multislice formulation, which takes thermal diffuse scattering fully into account. The improvement with regard to the classical frozen phonon approach is realized by separating the lattice configuration statistics from the dynamical scattering so as to avoid repetitive calculations. As an example, the influence of phonon scattering on the image contrast is calculated and investigated. STEM image simulation of crystals can be applied with reasonable computing times to problems involving a large number of atoms and thick or large supercells.

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1. Introduction

High-resolution transmission electron microscopy (HRTEM) has evolved into a very powerful tool to study the atomic structure in material science although it demands the use of modern computing methods and facilities. Experimental work is complemented by detailed simulations that capture the interaction of the electron with the specimen and the image formation, including coherent and incoherent aberrations. These calculations are based on a full quantum mechanical treatment of the dynamical scattering that occurs during electron propagation through the specimen. Such simulations are needed to support the interpretation of experimental data as well as to extract and explain additional information from the recorded micrographs.

Imaging in scanning transmission electron microscopy (STEM) with a high-angle annular dark field (HAADF) detector in a low-indexed zone axis directly gives the structure of the illuminated specimen free of contrast reversals over a large range of thicknesses, thus overcoming problems caused by dynamical diffraction in other imaging modes [1–4]. Moreover, the recorded micrographs are mainly incoherent and exhibit strong intensity dependence on atomic number [5,6], which is the key idea of HAADF STEM imaging. Despite the ability to interpret the structure directly, quantitative analysis of HAADF STEM requires simulation of both coherent Bragg reflections [7] and incoherent thermal diffuse scattering (TDS) [8] in a crystal.

In the past, several approaches have been developed to simulate the image formation in STEM with inclusion of thermal diffuse scattering (quasi-elastic) and inelastic scattering. In the “frozen phonon” model, first proposed in Ref. [9], a number of calculations is performed for various static configurations of atoms, slightly displaced from their mean positions. Then the image intensity is calculated by averaging the intensities obtained from different lattice configurations. According to Ref. [10], this method can be considered as the most accurate one, although it is very time consuming. Wang and Cowley
proposed a method based on the multislice formalism, which includes TDS within the first order approximation [10–12]. Dinges and Rose developed a semi-analytical approach within the multislice formalism [13]. Allen and Rossouw [14], Pennycook and Jesson [8] and Watanabe et al. [15] have incorporated TDS in the Bloch wave formalism. Allen et al. [16,17] have modified the Bloch wave theory to account for lattice-resolution contrast derived from both coherent and incoherent scattering mechanisms. TDS has been included through the use of a mixed dynamic form factor (MDFF) that can be approximated to a local scattering potential. An equivalent method amenable to calculate TDS via multislice techniques is presented there as well. The MDFF for the Einstein approximation of TDS is given in Ref. [18]. All these approaches assume the Einstein approximation for the atom vibrations. Muller et al. went beyond the Einstein approximation and simulated TDS including a detailed phonon dispersion curve [19].

In this paper we have implemented a method for HAADF STEM image calculations based on the real-space multislice theory of TDS introduced by Fanidis et al., where the lattice configuration average is performed independent of the time-varying simulations of the dynamical scattering [20]. As an example, we apply it to study the role of thermal diffuse scattering in HAADF STEM since in this imaging technique the role of TDS is crucial. The validity and accuracy of the method are proven by comparison of the results based on the models with two generation functions with those based on the frozen phonon model.

2. Model

There are two main approaches for the calculation of atomic-resolution STEM images. One of them is the Bloch wave method [5,15,22] proposed by Bethe in Ref. [23]. The other approach is the multislice many-beam dynamic electron diffraction method of Cowley and Moodie [24]. In this work we use the second method to simulate dynamic electron scattering since we intend to investigate atomic level resolution images, which usually involve many diffracted beams. Furthermore, this method does not require three-dimensional periodicity of the crystal. Therefore, its application is more convenient to study defects in crystals, such as displacements of atom positions. Finally, the multislice method is numerically better suited for the calculation of thickness-dependent characteristics, because the wave propagation through the crystal can be calculated slice-by-slice.

1The electron–phonon scattering mechanism does not cause significant energy-loss of strongly accelerated electrons but produces large momentum transfer, which can scatter the electrons to the angular range of the ADF detector. Moreover, measurements on amorphous carbon made in Ref. [21] as a function of spatial frequency show that images have high contrast for low spatial frequencies but exhibit a strong and constant reduction of the contrast for frequencies above 0.5 nm\(^{-1}\).

2.1. Theoretical background

Originally the multislice method was derived from a physical optics approach under the small-angle-scattering approximation neglecting backscattering. It can also be deduced from Feynman’s path-integral formalism for high-energy electron diffraction [25,26] or directly from the Schrödinger equation [27]. The forward-scattering assumption for high-energy electrons is investigated and validated in Ref. [28].

In the multislice method the crystal is divided into many parallel slices. If the thickness of each slice is sufficiently small, the scattering of each slice can be based on the projected potential approximation [29,30]. Labeling the wave function at the top of each slice by \( \Psi_n(p = (x, y)) \) and the propagator and transmission function for each slice by \( P_n(p, \Delta z_n) \) and \( T_n^0(p) \), respectively, the basic first-order multislice equation, which describes the diffraction of the electron beam by the crystal as a consecutive slice-by-slice process up to the exit face of the specimen, can be written as [31,32]:

\[
\Psi_{n+1}(p) = P_n(p, \Delta z_n/2) \otimes \{ T_n^0(p)[P_n(p, \Delta z_n/2) \otimes \Psi_n(p)]\} + \mathcal{C}(\Delta z^2),
\]

(1)

where

\[
P_n(p, \Delta z_n) = \frac{k_z}{i \Delta z_n} \exp \left[ \frac{i \pi k p^2}{\Delta z_n} \right].
\]

(2)

\[
T_n^0(p) = \exp \left[ i \sigma \frac{k_z}{k} \int_{z_n}^{z_{n+1}} V_n(p, z) \, dz \right]
\]

\[
=: \exp[i \sigma U_n(p)],
\]

(3)

with \( \Delta z_n \) the thickness of the \( n \)th slice, \( V_n(p, z) \) the potential field in the \( n \)th slice and \( \sigma = e/hv \) an interaction parameter with \( v \) the velocity of incident electron. It is possible to develop a second-order multislice theory by introducing potential eccentricity as described in [32,33], but in the present work the \( z \)-dependence of the potential (for HOLZ) and the high accuracy of calculations are ensured by the small slice thickness.

The multislice theory is employed to simulate the HAADF STEM images [34,35]. In a STEM, the objective lens forms a narrow probe wavefunction \( \Psi_p(p, \rho_p) \) upon the specimen surface at position \( \rho_p \). A focused probe is usually calculated by integrating the aberration function over the objective aperture as [29]:

\[
\Psi_p(p, \rho_p) = A_p \int_{0}^{q_{\text{appr}}} \exp[-i\chi(q) - i2\pi q \cdot (p - \rho_p)] \, dq.
\]

(4)

where \( A_p \) is the normalization constant, \( q_{\text{appr}} \) is the maximum spacial frequency in the objective aperture, and \( \chi(q) \) is given by

\[
\chi(q) = \pi \lambda q^2 (0.5 C_5 \lambda^2 q^2 + \Delta f),
\]

(5)

with defocus \( \Delta f \), wavelength \( \lambda \), spatial frequency \( q \) in the image plane, and spherical aberration \( C_5 \). The probe is
scanned across the sample and the electron intensity scattered to the angular range of the ring-shaped detector forms a convergent beam diffraction pattern (see Fig. 1). The integration of the signal intensity as registered by the detector gives the image intensity at one point in the image. The detector function is written as

\[ D(q) = \begin{cases} 
1, & q \in \text{detector}, \\
0, & q \notin \text{detector}.
\end{cases} \tag{6} \]

Electron–phonon (thermal diffuse) scattering is caused by thermal vibrations of the atoms (nuclei). The time necessary for high-energy electrons to pass through the specimen is about two orders of magnitude smaller than the atomic vibration period. On the other hand, the atomic vibration period is much shorter than the time-interval between incoming electrons for a typical beam current. Therefore, vibrating atoms of the specimen are seen as being stationary for each incoming electron. Since the energy loss of incoming electrons due to the electron–phonon scattering is unobservably small \((\sim k_B T \approx 25 \text{ meV})\) compared with their kinetic energy, thermal-diffuse scattering can be considered as a statically averaged quasi-elastic scattering from the crystal with various configurations of static displaced atoms. This idea forms the basis of so-called “frozen” lattice model, which assumes that the entire crystal in one vibration configuration is a frozen lattice and the diffuse scattering generated in the whole crystal is quasi-elastic. The incoherency is introduced only at the last stage when intensities contributed by different lattice configurations are summed up incoherently. Based on a rigorous quantum mechanical phonon excitation theory, it has been proven in Ref. [10] that this model is an excellent approximation.

In the present work, TDS has been included also by the method as presented in [13,20,37,38], where the lattice configuration averaging is performed separately from the multislice propagation. As usual, in the treatment of scattering by atomic vibrations it is assumed that TDS is not accompanied by the excitation of the electronic states in the solid and the Born–Oppenheimer approximation is adopted. Under the high-energy approximation and ignoring the effect of multiple electron–phonon scattering in not-too-thick samples (first-order approximation) the wavefunction, describing the inelastically scattered electrons (fluctuating part of the crystal wavefunction) can be written as follows [37]:

\[ \delta \psi(\rho, z, t) = i\sigma \int_0^z d' O_p(\rho, z - z') [\delta V(\rho, z', t) \psi_0(\rho, z')] \]

\[ \tag{7} \]

where the perturbation potential \(\delta V(\rho, z, t) = \sum_i \delta V_{i,k}(\rho - \rho_{z,k}, z - z_k, t)\) is a sum of the TDS potentials of individual atoms in a crystal. Here \(\delta V_{i,k}(\rho - \rho_{z,k}, z - z_k, t)\) is the deviation of the instantaneous crystal potential seen by an electron at the time \(t\) from its equilibrium value. The independent of the crystal state function \(\psi_0(\rho, z')\) is the averaged electron wave function component. The kernel \(O_p(\rho, z - z')\) is a dynamic scattering operator and it can be interpreted as follows: at depth \(z' < z\) the electron wave is inelastically scattered due to the TDS generation potential \(\delta V(\rho, z', t)\), i.e. inelastically scattered part of the electron wavefunction \(\delta V(\rho, z', t) \psi_0(\rho, z')\), and is further dynamically diffracted from \(z'\) to \(z\) by the operator \(O_p(\rho, z - z')\) [37,40]. This operator dramatically increases the calculation time in the case of TDS compared to those without inelastic scattering. The operator can be neglected when the detected signal is given by integration of intensities over a wide-angle annular detector, since it only redistributes all the inelastically scattered electrons over the detector [37]. This assumption is only valid for HAADF STEM simulations.

2.2. TDS generation function

The intensity distribution of the electron wavefunction in the diffraction plane is just the sum of the intensities of the inelastically (TDS wavefunction) and elastically (Bragg reflections) scattered electrons, if the exposure times are large (incoherent summation). We obtain

\[ I(q, d, \rho_p) = |\psi_0(q, z = d, \rho_p)|^2 + |\delta \psi(q, z = d, \rho_p)|^2, \tag{8} \]

where \(d\) is the specimen thickness and \(\rho_p\) is a probe position. It is shown in Refs. [36,41] that the TDS potential \(\delta V(\mathbf{r}', t)\) is of the same magnitude as the atomic potential and is considerably narrower than the averaged value of the atomic potential \(V_0(\mathbf{r})\), indicating that TDS is a localized scattering process. Therefore, the intensity of thermal diffuse scattered electrons can be written in the
following form [37]:

\[
I_{\text{TDS}}(d, \rho_k) \approx \sigma^2 \sum_{k' k''} \int \int d\rho' d\rho'' D(\rho' - \rho'') \\
\times \left\langle \delta U_k(\rho' - \rho_k) \delta U_{k'}(\rho'' - \rho_{k''}) \right\rangle \\
\times \psi_0(\rho', z_k, \rho_k) \phi_0(\rho'', z_{k''}, \rho_{k''}),
\]

(9)

where \( \delta U_k(\rho - \rho_k) = \int \delta V(\rho - \rho_k, z, z_k, t) dz \). Here the time dependence has been dropped, because the vibration motion of the atoms is slow compared with the velocity of the incident electron, and the brackets \( \langle \cdots \rangle \) mean averaging over the statistical ensemble. The kernel operator \( \langle \delta U_k(\rho' - \rho_k) \delta U_{k'}(\rho'' - \rho_{k''}) \rangle \) can be colligated to the Van Hove dynamic form factor [42]. The operator \( D(\rho' - \rho'') \) is the inverse Fourier transform of the detector function \( D(\mathbf{q}) \).

We adopt here the Einstein phonon model, i.e. there is no phase coupling between the atomic vibrations. This assumption can influence the final result due to the fact that it removes the effect of the coherence of the atom vibrations. In fact in Ref. [43] using the Debye model and the Warren approximation it has been shown that the longitudinal coherence length extends only over 3–4 interatomic distances. The transverse coherence is effectively destroyed by the ADF detector if

\[
x_{in} > 1.22 \frac{\lambda}{\Delta},
\]

(10)

where \( x_{in} \) is the inner cutoff of the ADF detector, and \( \Delta \) is the transverse coherent distance [44,45]. Because the localization length of \( \delta U_k(\rho - \rho_k) \) is much smaller than the interatomic distance (see Ref. [41]),

\[
\left\langle \delta U_k(\rho' - \rho_k) \delta U_{k'}(\rho'' - \rho_{k''}) \right\rangle \approx \left\langle \delta U_k(\rho' - \rho_k)^2 \right\rangle \delta_{k k'}.
\]

(11)

Hence the TDS electrons generated from atomic sites \( k' \) and \( k'' \) are incoherent. This assumption is correct if the ADF detector registers all the inelastic electrons: \( D(\rho' - \rho'') = \delta(\rho' - \rho'') \) and it is a valid approximation if the inner radius of the ADF is much larger than the Bragg angles around the central beam [11,12]. We can also write

\[
\left\langle \delta U_k(\rho - \rho_k)^2 \right\rangle = \left( U_k(\rho - \rho_k)^2 \right) - \left( U_k(\rho - \rho_k) \right)^2.
\]

(12)

Since each atom in our model is considered as an independent oscillator along the coordinate axes, averaging is based on the probability distribution, which can be factorized

\[
P(p_1, \ldots, p_N) = \prod_{k=1}^{N} P_k(p_k)
\]

\[
= \prod_{k=1}^{N} \frac{1}{\sigma_{a_k}^2} \exp \left[ -\frac{(\rho_k - \rho_{k0})^2}{\sigma_{a_k}^2} \right],
\]

(13)

where \( \sigma_{a_k}^2 \) is the two-dimensional mean vibration quadratic amplitude and \( \rho_{k0} \) is the average position of the \( k \)th atom [29]. This allows one to compute Eq. (9) numerically, since it is completely factorized

\[
I_{\text{TDS}}(d, \rho_k) \approx \sigma^2 \sum_{n} \int d\rho \sum_{k \in n} \langle \delta U_n(\rho - \rho_k)^2 \rangle |\psi_0(\rho, z_n, \rho_k)|^2
\]

\[
\approx \sigma^2 \sum_{n} \int d\mathbf{q} |g_n(\mathbf{q}) \otimes \psi_0(\mathbf{q}, z_n, \rho_k)|^2 D(\mathbf{q}),
\]

(14)

where

\[
g_n(\mathbf{q}) \equiv FT \left[ \sum_{k \in n} \sqrt{\langle \delta U_n(\rho - \rho_k)^2 \rangle} \right]
\]

(15)

is the TDS generation function. In this expression sum is over all atoms of the \( n \)th layer. The proposed approach of the calculation of the TDS intensity of the \( n \)th layer is therefore somehow similar to that proposed in Ref. [16], where the TDS intensity of any layer is given by an integral over the electron density in that layer \( |\psi_0(\rho, z_n, \rho_k)|^2 \) multiplied with the local inelastic potential. The role of the local TDS potential is fulfilled by the square of the modified TDS generation function, which takes into account the size of the ADF detector.

In the present work we propose two ways of constructing the TDS generation function. Since the mean vibration amplitude is very small as compared with the interatomic distance we can expand the TDS generation potential into a standard Taylor series

\[
\delta U_n(\rho - \rho_k) = (x_k - x_{k0}) \frac{\partial}{\partial x_k} U_n(\rho - \rho_k)|_{x_k=x_{k0}}
\]

\[
+ (y_k - y_{k0}) \frac{\partial}{\partial y_k} U_n(\rho - \rho_k)|_{y_k=y_{k0}}
\]

\[
+ O(\Delta p_k^2).
\]

(16)

This expansion is only valid if \( \rho \gg \rho_{k0} \), because in the close vicinity of \( \rho_{k0} \) the condition

\[
U_n(\rho - \rho_{k0}) \gg (x_k - x_{k0}) \frac{\partial}{\partial x_k} U_n(\rho - \rho_k)|_{x_k=x_{k0}}
\]

(17)

is violated. In order to overcome the problem of potential divergence in this expansion a small arbitrary shift in the derivative of the potential is introduced in such a way that we can safely neglect the higher order terms in the Taylor expansion. Therefore, Eq. (16) can be written as

\[
\delta U_n(\rho - \rho_k) = (x_k - x_{k0}) \frac{\partial}{\partial x_k} U_n(\rho - \rho_k)|_{x_k=x_k'}
\]

\[
+ (y_k - y_{k0}) \frac{\partial}{\partial y_k} U_n(\rho - \rho_k)|_{y_k=y_k'}.
\]

(18)

Assuming the \( x \) and \( y \) coordinates to be uncorrelated, one obtains

\[
\left\langle \delta U_n(\rho - \rho_k)^2 \right\rangle = \left\langle \left( (x_k - x_{k0}) \frac{\partial}{\partial x_k} U_n(\rho - \rho_k)|_{x_k=x_k'} \right)^2 \right\rangle
\]

\[
+ \left\langle \left( (y_k - y_{k0}) \frac{\partial}{\partial y_k} U_n(\rho - \rho_k)|_{y_k=y_k'} \right)^2 \right\rangle.
\]

(19)
from which
\[
\langle \delta U_n(\rho - \rho_k)^2 \rangle \approx \frac{a_k^2}{2} \left( \frac{\partial}{\partial \chi_k} U_n(\rho - \rho_k) \right)^2
\]
\[
+ \frac{a_k^2}{2} \left( \frac{\partial}{\partial y_k} U_n(\rho - \rho_k) \right)^2,
\]
where \( a_k^2 = \langle (x_k - x_{0k})^2 \rangle + \langle (y_k - y_{0k})^2 \rangle \). Using the cylindrical symmetry of the projected atom potential, we finally obtain the expression for the TDS generation function
\[
\mathcal{G}_n^A(\rho) = i \sum_{k \in n} \sqrt{a_k^2} \left( \frac{\partial}{\partial \chi_k} U_n(\rho - \rho_k) \right).
\]  
(21)
The second form of the TDS generation function can be obtained from Eq. (14)
\[
\mathcal{G}_n^B(\rho) = i \sum_{k \in n} \sqrt{\langle \delta U(\rho - \rho_k)^2 \rangle},
\]  
(22)
where the summation is restricted to the atoms within one specimen slice and the intensities obtained from different slices are added incoherently. The Fourier transform of the \( \mathcal{G}_n^A \) generation function of phonon scattering exhibits a closer behavior to that of the Fourier transform of a phonon perturbation potential derived in Ref. [46], than \( \mathcal{G}_n^B \) does.

Due to the electron–phonon scattering the intensity of the elastically scattered electrons is attenuated by the absorption potential [13,39]. Therefore, it is possible to write
\[
T_n(\rho) = \exp \left[ i \sigma \left( U_n(\rho) - \mu_n^{A,B}(\rho)/2 \right) \right],
\]  
(23)
where the averaged projected potential field in the \( n \)th slice is
\[
U_n(\rho) = \sum_{k \in n} U_n(\rho - \rho_k)
\]  
(24)
and the absorption functions for the \( n \)th slice in the case of the first and the second forms have been derived in a way presented in Ref. [13] and are given by
\[
\mu_n^A(\rho) = \sigma^2 \sum_{k \in n} a_k^2 \left( \frac{\partial}{\partial \chi_k} U_n(\rho - \rho_k) \right)^2
\]  
(25)
and
\[
\mu_n^B(\rho) = \sigma^2 \sum_{k \in n} a_k^2 \langle \delta U(\rho - \rho_k)^2 \rangle,
\]  
(26)
respectively. Finally, in the plane of the detector the contributions to the intensity from elastic scattering and inelastic scattering of all layers are added incoherently.

3. Results and discussion

For a numerical simulation, Au- and Cu-crystal samples have been constructed with the following geometric parameters: the transversal size is \( 5a_0 \times 5b_0 \) (\( a_0 \) and \( b_0 \) are the in-plane lattice parameters perpendicular to the optical axis). This real space grid of 1024 \( \times \) 1024 pixels gives a real space sampling resolution equal to \( \Delta x \approx 0.2 \) Å/pixel. The resolution in reciprocal space is equal to \( \Delta q \approx 0.05 \) Å\(^{-1}\), and the angular resolution is equal to \( \Delta \theta = \lambda \Delta q \approx 0.98 \) mrad. This sampling restricts the calculations to a maximum detector angle of 340 mrad, which is adequate for the calculations [30]. The slice thickness is chosen as \( \epsilon_0/4 \). The phase-grating function [see Eq. (3)] is calculated for each slice within the unit cell and then stored. STEM line images are formed by scanning a narrow electron probe along a line through the center positions of the atomic columns. The probe is formed with an objective aperture radius of 11.2 mrad and the spherical aberration coefficient \( C_5 \) is taken equal to 0.05 mm. The defocus is 115 Å (Scherzer focus) and the accelerating voltage \( V = 300 \) kV. The beam direction is chosen along the [001] zone axis. Different angular detection ranges of the ADF detector have been chosen as well. The three-dimensional atomic mean vibration amplitude \( (a_{0w}^2)^{1/2} \) is taken equal to 0.084 Å for the Au-crystal and \( (a_{0w}^2)^{1/2} \) is equal to 0.085 Å for the Cu-crystal [29,47]. For an isotropic distribution of the mean squared atomic deviations, there is a simple relation between the three-dimensional and the two-dimensional version: \( 3a_k^2 = 2a_{0w}^2 \).

The atomic scattering factor is used in a parameterized form taken from Ref. [30]:
\[
f_e(q) = \sum_{i=1}^{3} \frac{a_i}{q^2 + b_i} + \sum_{i=1}^{3} c_i \exp(-d_i q^2),
\]
where the \( a_i, b_i, c_i \) and \( d_i \) coefficients are tabulated in Ref. [30] and which have the appropriate form at large and small angles.

Fig. 2 illustrates the thickness dependence of the simulated HAADF STEM intensity of a [001]-oriented Cu-crystal for the detector with annuli (a) 20–200 mrad and (b) 40–200 mrad, when the probe is located at the copper atom column. For both geometries of the detector it is obvious that the fraction of the image intensity that results from elastic scattering rapidly decreases with increasing thickness, while the total intensity (elastic plus inelastic, caused by thermal diffuse scattering) increases with thickness. For example, in the case of the inner detector radius of 20 mrad, at a crystal thickness of 40 Å, more than 60% of the image intensity results from purely elastic scattering, while at a thickness of 400 Å less than 10% originates from elastic scattering. In the former case, due to a stronger contribution of the elastic component the fringes in the total intensity curve are more pronounced. This figure shows the results of calculations based on two model generation functions for the total intensity. The curve marked as “Model A” has been calculated using the generation function from Eq. (21), while that marked as “Model B” has been based on Eq. (22). The function \( \mathcal{G}_n^A(\rho) \) generates stronger scattering into both angular ranges...
presented in the figure, while $\omega_B(\rho)$ generates less intensity especially in the case of the Au sample where the total signal occurs for smaller thicknesses. This is due to stronger absorption. The oscillatory behavior of the intensity is now almost suppressed and the intensity is a monotonic function of sample thickness. Again, the calculations based on model A give higher intensity than those based on model B. This figure also illustrates the results from rigorous calculations based on the frozen phonon model. One can see that the result based on model A is much closer to that of the frozen phonon model compared to model B. Assuming the frozen phonon model as a reference method, we can therefore conclude that model A is an adequate approach to simulate TDS.

Figs. 4a–d show the image contrast

$$\eta = \frac{I_{\text{max}} - I_{\text{min}}}{I_{\text{max}} + I_{\text{min}}}$$

as a function of sample thickness for copper, gold, and silicon crystals. Here $I_{\text{max}}$ and $I_{\text{min}}$ are the maximal and minimal intensities in the line scan of an image. Figs. 4a–c are presented here in order to show how thermal diffusion scattering disturbs the image contrast. One can infer, that in both structures (copper and gold) the inclusion of thermal diffuse scattering in the calculations results in a strong decrease of the contrast with sample thickness. However, this contrast loss heavily depends on the “weight” of the sample, i.e. on the atomic number of the atoms in the sample. At the same crystal thickness the reduction in image contrast is larger for the high-Z elements than for low-Z elements. From the analysis of the panels (a) and (b) of the figure it is possible to conclude that an increase of the inner radius of the detector leads to a stronger influence of TDS on the image contrast. This is in conformity with the fact that at higher scattering angles

Fig. 2. Elastic and total (inelastic plus elastic) intensities in the annular dark field detector with an angular range (a) 20–200 mrad; (b) 40–200 mrad for a probe located over a Cu column as a function of sample thickness.

Fig. 3. Elastic and total (inelastic plus elastic) beams falling on the annular dark field detector with an angular range 40–200 mrad as a function of sample thickness.
the TDS waves make the main contribution to the image contrast. From panel (b) one can see, that in the case of the detector with annulus 40–200 mrad at 300 Å copper sample thickness, TDS reduces the contrast from $\eta = 0.66$ to $\eta = 0.45$ (Model B) or even to $\eta = 0.38$ (frozen phonon model). At a thickness $d = 500$ Å the contrast decreases to $\eta = 0.39$ (Model B) or even to $\eta = 0.31$ (frozen phonon model). In the case of a gold sample (panel c)) with thickness $d = 500$ Å the contrast factor, which can be defined as $F_{\text{contrast}} = \eta_{\text{without TDS}} / \eta_{\text{with TDS}}$, equals to 3.75 (Model B) or even more than 5 (frozen phonon model). The results obtained from the frozen phonon model are always slightly lower than those based on models A and B. This observation can be explained by the fact that calculations based on the frozen phonon approximation are not restricted to single thermal diffuse scattering, whereas models A and B are. However, this assumption is valid for not-too-thick samples as can be seen from Fig. 4.

Fig. 4d is introduced in the present paper in order to compare the results presented in Ref. [38] with those obtained by applying models A and B to a [011]-oriented Si crystal. In these calculations a 100 kV STEM has been assumed with $C_s = 0.05$ mm, $\Delta f = 136$ Å and an aperture radius of 24.9 mrad. An annular detector with limiting angles $\alpha_{\text{in}} = 50$ mrad, $\alpha_{\text{out}} = 200$ mrad has
been chosen to be consistent with the paper of Dinges and Rose [38]. The three-dimensional mean vibration amplitude $a_{Si}$ is equal to 0.076 Å. From Fig. 4d one can see that the contrast calculated using the frozen phonon model, models A and B as well as results of Ref. [38] show a decreasing trend with small oscillations with increasing thickness. Although this is in agreement with the results obtained in Ref. [38], a discrepancy of the order of 15% is found between the results calculated in this study and those taken from [38].

4. Conclusions

In order to investigate the influence of the phonon scattering on the image contrast in STEM mode we have developed a model for numerical calculation of the coherent elastic scattering and the incoherent TDS. The method reduces the computing time for a HAADF STEM image calculation by a factor of 3–5 as compared with that required by a frozen phonon model and it gives very reasonable results if one is interested in the relative intensity characteristics of the simulated images. The simulations show that inclusion of TDS in the image simulation leads to a strong reduction of the image contrast in HAADF STEM. The loss of the contrast depends on the “weight” of the sample, the sample thickness and on the geometry of the detector in the case of a STEM imaging and varies from less than 2 to almost 5. Although the present work focuses on STEM, it is clear that the influence of TDS is also present in HRTEM. Therefore, it is likely that TDS makes the main contribution to the Stobbs factor—the contrast discrepancy between experimental and simulated images.

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