Precision of three-dimensional atomic scale measurements from HRTEM images: What are the limits?

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1. Introduction

Structural information including atom column positions, mass of atom columns and surface profile is embedded in image data recorded by means of high resolution transmission electron microscopy (HRTEM). The ultimate goal of HRTEM is to determine this information not only qualitatively but also quantitatively. In other words, the atom column positions, the mass of atom columns and the surface profile should be measured as precisely as possible on a local atomic scale. In order to reach this goal, statistical parameter estimation theory has proven to be very effective. This theory makes use of a model simulating the electron–object interaction, the microscope transfer, and image detection. The model describes the expectations of the image pixel values and contains a set of parameters that have to be measured. These parameters represent the structural information and they are estimated by fitting the model to the experimental data using a goodness-of-fit criterion, such as the likelihood function. In practice, atom column positions can be estimated with a precision in the picometer range using statistical parameter estimation theory.

Despite the high precision that can nowadays be achieved for the atom column positions [2–7], the atom column mass and surface profile cannot routinely be measured at the atomic scale using HRTEM. One of the problems is that, at the atomic scale, the surface is never flat due to the relative protrusion of the atom columns compared with neighboring columns [8]. This protrusion is caused by the fact that neighboring atom columns have different numbers of atoms and/or are at different vertical positions. Therefore, the distance between the exact exit position of the atom column and the image plane is different for every atom column. This offset is called the defocus. Measuring the defocus value for every atom column would reveal the surface profile of the object. The purpose of this study is to understand the theoretical limits with which one can measure the mass and surface profile atom column by atom column. Ultimately, the reliability with which one can measure these parameters is only limited by the unavoidable presence of noise in the observations. This sets limits to the precision of the parameter estimates that can be obtained. Since the mass is proportional to the thickness provided the atom column type and the interatomic distance are known, we compute the highest attainable theoretical precision with which the thickness and the surface profile can be estimated. This allows us to analyze under which conditions atomic scale measurements can be obtained and to explore the limits on the precision. In this paper, we consider two types of
atom columns: Au[100], representing a heavy atom column type, and Si[100], representing a light atom column type.

This paper is organized as follows. First, we derive a parametric model describing the expectations of the observations, the image pixel values, in Section 2. In Section 3, given the model and knowledge about the statistical distribution of the observations, we explain how the Fisher information [9] results in an expression for the attainable precision with which structure parameters can be estimated. This expression, which is called the Cramér–Rao lower bound (CRLB), is a function of the object parameters, the microscope parameters, and the electron dose. Next, we use the CRLB to determine the optimal microscope settings in terms of the attainable precision with which structure parameters can be estimated, using the principles of Bayesian statistical experimental design [10]. These settings include the spherical aberration constant, the chromatic aberration constant and the defocus. Finally, in Section 4, we explore the limits of the estimation precision for thickness and surface profile under optimal microscope settings and realistic settings for the electron dose. In Section 5, we draw some conclusions.

2. Image model

In this section, we derive a parametric model that describes the expectations of the intensity recorded by the HRTEM. This model contains microscope parameters, such as objective aperture, spherical aberration constant, defocus and electron dose, and object parameters, such as the positions, thicknesses and widths of the atom columns. The model derivation involves three steps. First, we describe the dynamical scattering between the electron beam and the object resulting in the complex wave function at the exit plane of the object. To this end, we make use of the channelling theory that provides a parametric expression for the exit wave, i.e. the electron wave at the exit plane of the object as the electrons pass through the atom columns. Second, we include the transfer function of the microscope to derive the image wave. Third, we derive an expression for the image intensity distribution and the expectation model.

2.1. The exit wave

If the object is orientated along a main zone axis and provided that the distance between adjacent columns is not too small, the dynamical interaction between the electron beam and the object can be described by means of the channelling theory [11,12]. The complex wave function at the plane where the electron beam leaves the object is called the exit wave. The channelling theory assumes that the exit wave, which we denote by \( \psi_c \), can be expressed in terms of the lowest bound state of the atom columns only [13]:

\[
\psi_c(r,z) = 1 + \sum_{n=1}^{N_c} c_n \phi_{s,n}(r-b_n) \left[ \exp \left( -i \frac{E_{s,n}}{E_0} r \right) - 1 \right],
\]

(1)

where 1 corresponds to an incident plane wave, \( E_0 \) is the incident beam energy, \( \lambda \) is the wavelength, \( r \) is the two-dimensional vector in the plane perpendicular to the incident beam direction, and \( N_c \) is the total number of atom columns. The function \( \phi_{s,n}(r-b_n) \) is the s-state wave function of the nth atom column at position \( b_n = (b_{nx}, b_{ny}) \) with energy \( E_{s,n} \) that depends on the type of atom column and the interatomic distance. The product of \( E_{s,n} \) and the atom column thickness \( z_n \) is the total mass of the atom column. The s-state wave function may be approximated by a quadratically normalized and parameterized Gaussian function with column-dependent width \( a_n \) [13],

\[
\phi_{s,n}(r) = \frac{1}{a_n \sqrt{2\pi}} \exp \left( -\frac{r^2}{4a_n^2} \right),
\]

(2)

where \( r = |r| \). The excitation coefficient \( c_n \) is \( c_n = 2\sqrt{2\pi a_n} \).

From Eq. (1), it follows that the electron wave oscillates periodically inside the atom column. The oscillation distance \( z_{ext,n} \) is called the extinction distance [14] and is a function of the average mass density of the column:

\[
z_{ext,n} = \frac{2E_0}{E_{s,n}} \frac{1}{\lambda_c}.
\]

(3)

The wave function in the image plane \( \psi_t \) can be written as the convolution product of the exit wave and the point spread function \( t(r) \) of the electron microscope [15]:

\[
\psi_t(r,x) = \psi_c(r,z) \ast t(r).
\]

(5)

The Fourier transform of \( t(r) \) represents the microscope transfer function \( T(g) \) with \( g \) the spatial frequency vector in Fourier space. In what follows, we describe the image formation mathematically including the damping effect due to partial spatial and temporal coherence and lens aberration effects resulting from predominantly determined aberrations that are aligned with the optic axis. Under these conditions the function \( T(g) \) is radially symmetric [15] in the spatial frequency range such that \( T(g) = T(g) \). The transfer function can be described as a product of the circular aperture function \( A(g) \), the damping envelope \( D(g) \) and the phase shift \( \chi(g) \) resulting from the objective lens aberrations:

\[
T(g) = A(g) \bar{D}(g) \exp(-i\chi(g)),
\]

(6)

where the aperture function \( A(g) \) is defined as

\[
A(g) = \begin{cases} 
1 & \text{if } g \leq g_{ap}, \\
0 & \text{if } g > g_{ap},
\end{cases}
\]

(7)

with \( g_{ap} \) the aperture radius. In this study, we assume that there is no objective aperture so that \( A(g) \) is constant and equal to 1. The damping envelope \( D(g) \) describes the incoherent effects including partial spatial coherence [16,17] and temporal coherence [18]:

\[
D(g) = \exp \left( -\frac{\pi^2 (\lambda C_\epsilon \lambda^2 g^2 + \pi g^2)^2}{\ln 2} \right) \exp \left( -\frac{\pi^2 \lambda^2 A^2 g^2}{2} \right),
\]

(8)

with \( \lambda_c \) the semi-angle of beam convergence, \( C_\epsilon \) the spherical aberration constant, \( \epsilon \) the defocus and \( A \) the defocus spread due to chromatic aberration. The defocus spread \( A \) is defined as [19]

\[
A = C_\epsilon \sqrt{\frac{4 \left( \frac{\Delta E}{E_0} \right)^2 + \left( \frac{\Delta V}{V_0} \right)^2 + \left( \frac{\Delta \lambda}{\lambda} \right)^2}{\lambda}}.
\]

(9)

with \( C_\epsilon \) the chromatic aberration constant, \( \Delta I \), \( \Delta V \) and \( \Delta \lambda \) the standard deviations of the objective lens current \( I_0 \), the accelerating voltage \( V_0 \) and the incident electron energy \( E_0 \), respectively. In the following, we assume that \( \Delta V/\lambda \) and \( \Delta \lambda/E_0 \) are much smaller than \( \Delta E/E_0 \), such that Eq. (9) reduces to

\[
A = C_\epsilon \frac{\Delta E}{E_0}.
\]

(10)

This approximation is valid for a small effective incoherent electron source in case the central unscattered beam is much stronger than the scattered beams [16,17]. The phase shift \( \chi(g) \) is
given by
\[
\chi (g) = \pi e^2 k_0^2 + \frac{\pi C_1^2 g^2}{2}.
\]
(11)
Although other higher order aberrations could be included as well, they do not change the major conclusions of this paper. Therefore, we can ignore them in the remainder of this paper in order not to complicate the derivation of the optimal microscope settings unnecessarily.

2.3. The intensity distribution and image recording

The image intensity distribution is given by the modulus square of the image wave:
\[
l(r) = \left| \psi(r, z) \right|^2.
\]
(12)
From Eqs. (1) and (5), it follows that
\[
l(r) = \left| 1 + 2 \pi \sum_{n=1}^{N_t} C_n \left[ \exp\left(-i n \frac{E_{n}}{E_0} k z_n\right) - 1 \right] \right|^2 \times \int_0^\infty \phi_{\text{int}}(g) T(g) |2 \pi g (r - \beta_n)| g \, dg \]
(13)
in which the integral expression, representing the convolution product, follows from the fact that \( \phi_{\text{int}}(r) \) and \( T(r) \) are both radially symmetric. In this equation, \( J_n \) is the zeroth-order Bessel function of the first kind. Notice that the contrast of the image of a particular atom column also oscillates periodically with thickness due to the dynamical effects described in Eq. (4) in Section 2.1.

Next, we derive an expression describing the expected number of electrons recorded on a charge-coupled device (CCD) camera. The recording device is characterized by \( K \times L \) equidistant pixels of area \( \Delta x \times \Delta y \), where \( \Delta x \) and \( \Delta y \) are the sampling distances in the \( x \) and \( y \) directions, respectively. We choose the field of view (FOV) large enough so as to guarantee that all redistributed information from the tails of the point spread function \( t(r) \) of the microscope is collected. Furthermore, we assume that the quantum efficiency of the CCD camera is sufficient to detect single electrons and that there is no objective aperture, so that the total number of electrons leaving the electron source equals the number of detected electrons. According to [16], the total number of electrons is given by
\[
N_e = \frac{B_t E_a FOV \pi 2^2}{e^2},
\]
(14)
with \( t \) the recording time, \( B_t \) the reduced brightness and \( e = 1.6 \times 10^{-19} \text{ C} \) the electron charge. The expected number of electrons at pixel \((k,l)\) is then given by
\[
A_{kl} = N_e p_{kl},
\]
(15)
where \( p_{kl} \) is the probability that an electron hits the pixel \((k,l)\). The probability \( p_{kl} \) can be approximated as
\[
p_{kl} = \frac{l(k,l)}{\sum_{k,l} l(k,l)},
\]
(16)
provided \( \Delta x \) and \( \Delta y \) are small compared to the atom column width. Furthermore, \( l(k,l) \) is found by evaluating Eq. (12) at the pixel \((k,l)\) positioned at \( r = (x_k, y_l) \).

3. The Cramér–Rao lower bound

In the previous section, we derived a model describing the expectations of the observations, the image pixel values. This model depends on the set of parameters \( \theta \) including the positions \( \beta_n \) and thicknesses \( z_n \) of the atom columns. These are the parameters to be determined. If it were possible to describe the observations perfectly well by the model, we could find the parameters resulting in a perfect fit and the parameters would be measured exactly. However, despite technical progress, which provides us with detectors that are able to count single electrons, the pixel values can only be described in probability terms so that the images cannot be described exactly by the model. This is due to the inherent presence of noise in the images. In electron microscopy, the image pixel values are electron counting results which are binomially distributed. However, for a large electron dose \( N_e \) and small \( p_{kl} \), the binomial distribution can be approximated by the Poisson distribution [20,21]. In this study, we are mainly interested in the precision with which the thickness and surface profile can be estimated. To estimate these structure parameters, a maximum likelihood estimator can be used. The precision of such an estimator can be presented by the variance or the standard deviation, being the square root of the variance. In general, different estimators have different precisions. However, the variance of an unbiased estimator can never be lower than the so-called Cramér–Rao lower bound (CRLB). This lower bound thus represents the attainable precision. Fortunately, there exists a class of estimators, including the maximum likelihood estimator, that achieves the CRLB asymptotically, that is, for increasing number of observations. We have tested this by means of simulations (see Appendix). An expression for the CRLB can be derived using the concept of Fisher information [1].

The Fisher information contained within a set of observations can be derived from the expectation model derived in Section 2 and the joint probability density function of these observations. For independent Poisson distributed observations, the probability that a set of observations \( w = (w_{11}, w_{21}, \ldots, w_{KL}) \) is equal to \( \omega = (\omega_{11}, \omega_{21}, \ldots, \omega_{KL}) \) is given by
\[
P(\omega; \theta) = \prod_{k=1}^{K} \prod_{l=1}^{L} \frac{A_{kl}^{W_{kl}}}{\omega_{kl}^{W_{kl}}} \exp(-A_{kl}),
\]
(17)
with \( A_{kl} \) the expectation of the observation \( w_{kl} \). These expectations are described by the model in Eq. (15) containing the parameters \( \theta \) to be estimated. The Fisher information matrix \( F \) with respect to the elements of the \( M \times 1 \) parameter vector \( \theta \) is defined as the \( M \times M \) matrix
\[
F = -E \left[ \frac{\partial^2 \ln P(\omega; \theta)}{\partial \theta \partial \theta^T} \right],
\]
(18)
where \( E[\cdot] \) denotes the expectation operator and the superscript \( T \) denotes the transposition of the vector \( \theta \) [9]. The matrix inside the brackets represents the Hessian matrix of \( \ln P(\omega; \theta) \). From Eqs. (17) and (18), it follows that the \((r,s)\)th element of \( F \) is equal to
\[
F_{rs} = \sum_{k=1}^{K} \sum_{l=1}^{L} \frac{\partial A_{kl}}{\partial \theta_r} \frac{\partial A_{kl}}{\partial \theta_s}.
\]
(19)
It can be shown that the covariance matrix \( \text{cov}(\hat{\theta}) \) of any unbiased estimator \( \hat{\theta} \) is larger than or equal to the inverse of the Fisher information matrix \( F \) [22]:
\[
\text{cov}(\hat{\theta}) \geq F^{-1}.
\]
(20)
The matrix \( F^{-1} \) is called the Cramér–Rao lower bound on the variance of \( \theta \). This inequality expresses that the difference of the matrices \( \text{cov}(\hat{\theta}) \) and \( F^{-1} \) is positive semidefinite. Since the diagonal elements of \( \text{cov}(\theta) \) represent the variances of \( (\hat{\theta}_1, \ldots, \hat{\theta}_M) \), these variances are larger than or equal to the corresponding diagonal elements of \( F^{-1} \):
\[
\text{var}(\hat{\theta}_r) \geq [F^{-1}]_{rr},
\]
(21)
with \( \hat{\theta}_r \) the estimator of the \( r \)th parameter of the vector \( \hat{\theta} \).

In Section 4, we compute the CRLB which serves as the lower bound for the estimates of the thickness of the atom columns and the surface profile. We will use this lower bound to investigate
the theoretical limits on the precision with which the thickness and surface profile of an individual atom column can be estimated. In [20,23–27], other examples are shown in which the CRLB has been used to optimize the microscope design in the context of atom column position measurements.

4. Results and discussions

In this section, we use the CRLB, as described in Section 3, to quantify the maximum attainable variance when estimating the thickness and surface profile of an atom column. We consider two types of atom columns, Si[100] and Au[100] representing light and heavy columns, respectively. First, we choose a criterion to optimize the microscope settings. Next, we provide an overview of the microscope settings, and optimize a selection of them. Finally, we present numerical results for Si and Au atom columns with and without neighboring atom columns.

4.1. Optimality criterion

The first step of this study is to find the microscope settings with which the unknown object parameters can be measured as precisely as possible. Although we are mainly interested in exploring the limits with which the thickness and surface profile can be estimated, we have to take into account the positions of atom columns. Indeed, we cannot estimate the thickness of an atom column without knowing the positions of the atom columns. Therefore, the optimization of the microscope design has to include all object parameters simultaneously. As shown in Section 3, the smallest possible variance for a certain parameter estimate is given by the corresponding diagonal element of the CRLB. Unfortunately, simultaneous minimization of these diagonal elements as a function of the microscope parameters is usually impossible. A decrease of one diagonal element often leads to an increase of another diagonal element. Therefore, a summary measure, that gives a trade-off between the variances of all parameter estimates, is needed. One such summary measure is the A-optimality criterion [28] which we used in this study. The A-optimality criterion is defined as the trace of the CRLB, that is, the sum of the diagonal elements of the CRLB:

$$\text{tr}(\text{CRLB}) = \sum_{r=1}^{M} (F^{-1})_{rr}. \quad (22)$$

Minimizing the A-optimality criterion with respect to the microscope settings thus yields the experimental design that, on average, results in the most precise estimates of the object parameters $\theta$.

However, the optimal design depends on the values for the object parameters including the column thickness. We now face the problem that, in general, the thickness is not known a priori but needs to be estimated to the contrary. We circumvent this problem that, in general, the thickness is not known a priori but needs to be estimated to the contrary. We circumvent this problem by choosing a discrete uniform prior distribution for the thickness, such that we obtain the minimal possible variance for a certain parameter estimate. As we have no prior knowledge about the thickness, we choose a discrete uniform prior distribution for the thickness, such that we obtain the following Bayesian A-optimality criterion:

$$\sigma_B^2 = \sum_{z=1}^{d} \text{tr}(\text{CRLB}), \quad (23)$$

where $z$ is the thickness which in principle takes values from $1 \cdot d$, thickness in case of 1 atom, up to $N_{\text{m}} \cdot d$, corresponding to the thickness of an infinite number of atoms. Here, $d$ corresponds to the interatomic distance. However, since the expectation model oscillates periodically with thickness as described in Section 2.3, the value of $\sigma_B^2$ also oscillates periodically with thickness. Therefore, we only need to sum over thickness values within one extinction distance. The microscope settings for which $\sigma_B^2$ is minimal then form the optimal design.

4.2. Microscope parameters

An overview of the fixed microscope parameters that enter the parametric model discussed in Section 2 is given in Table 1. The parameters include the electron energy $E_0$, the semi-angle of beam convergence $\zeta$, and the reduced brightness $B_r$. Parameters that describe the image recording are also given, including the recording time $t$, and the number of pixels $K$ and $L$ in the $x$- and $y$-directions with pixel sizes, $\Delta x$ and $\Delta y$, respectively. The $\text{FOV}$ is chosen wide enough to include the tails of the microscope transfer function. Using Eq. (14), the electron dose corresponds to about $1.2 \times 10^4 \text{ counts/A}^2$ for the parameters given in Table 1. For simplicity, the parameters given here are kept constant throughout the study. Other microscope parameters such as the spherical aberration constant $C_s$, the chromatic aberration constant $C_c$ and the defocus $\epsilon$ are optimized by minimizing the Bayesian A-optimality criterion $\sigma_B^2$.

4.3. Numerical results

In this subsection, we present the results of the numerical evaluation of the CRLB on the thickness. First, we consider an isolated atom column. Next, we take into account overlap of neighboring columns. In Table 2, the structure parameters are given for two types of atom columns used in this study, Au[100] and Si[100]. These include the energy of the s-state $E_s$, the width of the atom column $a$, the interatomic distance $d$ in the atom column and the extinction distance $z_{\text{ext}}$ in [100] orientation (see [14,29]).

4.3.1. Isolated atom columns

For an isolated atom column, the parameter vector $\theta$ includes the positions and the thickness of the atom column, such that $\theta=(\beta_x, \beta_y, \alpha, z)$. The atom column is chosen to be located at the center of the $\text{FOV}$, such that $(\beta_x, \beta_y) = (0, 0)$. Thus, the CRLB is a $3 \times 3$ matrix of which the diagonal elements are the lower bounds on the variances of the estimates of $\beta_x$, $\beta_y$ and $z$, respectively.

First, the minimum of $\sigma_B^2$ should be found so that the microscope settings leading to the most precise object parameter estimates can be obtained. Finding the optimal settings of the microscope requires solving a non-linear minimization problem. In general, this can be done using an iterative optimization

<table>
<thead>
<tr>
<th>Table 1</th>
<th>Microscope parameters.</th>
</tr>
</thead>
<tbody>
<tr>
<td>$E_0$</td>
<td>$10^4 \text{ rad}$</td>
</tr>
<tr>
<td>$\zeta$</td>
<td>$2 \times 10^7 \text{ Am}^{-2} \text{s}^{-1} \text{V}^{-1}$</td>
</tr>
<tr>
<td>$t$</td>
<td>1 s</td>
</tr>
<tr>
<td>$K$</td>
<td>129</td>
</tr>
<tr>
<td>$L$</td>
<td>129</td>
</tr>
<tr>
<td>$\Delta x$</td>
<td>0.2 Å</td>
</tr>
<tr>
<td>$\Delta y$</td>
<td>0.2 Å</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Table 2</th>
<th>Structure parameters of Au[100] and Si[100] atom column types.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Column type</td>
<td>$E_s$ (eV)</td>
</tr>
<tr>
<td>Au[100]</td>
<td>−210.8</td>
</tr>
<tr>
<td>Si[100]</td>
<td>−20.2</td>
</tr>
</tbody>
</table>
method requiring proper starting values. However, in our study, the number of microscope settings is limited. Therefore, we computed the Bayesian A-optimality criterion value $\sigma_B^2$ for all possible combinations of the spherical aberration constant $C_s$, the chromatic aberration constant $C_c$ and the defocus $\epsilon$. We considered values for $C_s$, $C_c$ and $\epsilon$ within a range which is generally used in electron microscopy. These optimal settings we obtained are listed in Table 3. To illustrate this, Fig. 1 shows $\sigma_B^2$ as a function of the spherical aberration constant $C_s$ and the defocus $\epsilon$ at $C_c = 0$ mm. All values in Fig. 1 represent the actual values except at $C_s = 0$ mm and $\epsilon = 0$ Å where the value for $\sigma_B^2$ is much larger. This value has been reduced for reasons of visualization only. The reason why $C_s = 0$ mm and $\epsilon = 0$ Å are not optimal is that, for a distance close to the extinction distance, the contrast vanishes almost completely. This leads to excessive values for the variances of the estimates of the position coordinates, and, hence, to a large value for $\sigma_B^2$. Examples of the standard deviation of the thickness estimator as a function of $C_s$ and $C_c$ for thicknesses of a half and one extinction distance are given in Fig. 2 for a Au[100]

<table>
<thead>
<tr>
<th>Column type</th>
<th>$C_s$ (mm)</th>
<th>$C_c$ (mm)</th>
<th>$\epsilon$ (Å)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Au[100]</td>
<td>0</td>
<td>0</td>
<td>50</td>
</tr>
<tr>
<td>Si[100]</td>
<td>0.2</td>
<td>0</td>
<td>-700</td>
</tr>
</tbody>
</table>

Fig. 1. Plots of $\sigma_B^2$ as a function of spherical aberration constant $C_s$ and defocus $\epsilon$ for isolated (a) Au[100] and (b) Si[100] atom columns. The chromatic aberration constant $C_c$ is set equal to 0 mm.

Fig. 2. The standard deviation of the thickness estimate (a) as a function of spherical aberration constant $C_s$ with $C_c$ and $\epsilon$ at the optimal settings given in Table 3 and (b) as a function of chromatic aberration constant $C_c$ with $C_s$ and $\epsilon$ at the optimal settings given in Table 3 for an isolated Au[100] atom column. The thickness corresponds to half an extinction distance • and to one extinction distance ▲.
atom column. This figure shows, for example, that at a thickness of half an extinction distance, the standard deviation can be improved by a factor of about 1.5 by reducing the spherical aberration constant from 0.4 mm to 0 mm. By reducing the chromatic aberration constant from 1.2 mm to 0 mm the improvement is a factor of 1.4. Throughout this paper, we use the microscope parameters given in Tables 1 and 3. Comparing the diagonal elements of the CRLB shows that the trace of the CRLB is dominated by the variance of the thickness estimator. The lower bound on the standard deviation of the thickness estimate is 30 times larger than the standard deviation of the estimates of the position coordinates for Au[100] and 80 times larger than those for Si[100]. This implies that the optimal settings are mainly determined by the minimization of the variance of the thickness. As a result, acquiring three-dimensional structure information in terms of thickness is much more difficult than obtaining two-dimensional information on the projected atom column positions.

In order to determine the number of atoms in a column, or, in other words, the thickness, from HRTEM images, exit wave reconstruction is often applied. In order to reconstruct the complex exit wave, a series of images at different defocus values is recorded [30–32]. We have therefore investigated under which conditions the image series should be recorded so as to estimate the thickness as precisely as possible. In particular, we have studied the required number of images. In this optimization, we assume that the total number of electrons interacting with the object is limited to avoid radiation damage. Therefore, we have assumed a constant electron dose which is equally distributed over the images of the series. Fig. 3 shows the lower bound on the standard deviation with which the thickness can be estimated as a function of the number of images for a Au[100] column at optimal microscope settings. The defocus step between the images in a series is 50 Å. From this figure, it can be seen that the standard deviation is minimum if only one image is recorded. The precision does not improve by taking more images. The reason for this is that the number of electrons per image decreases with increasing number of images, which entails a decrease in the signal-to-noise ratio. The most precise estimate for the thickness is thus obtained when all electrons are used to form a single image at the optimal defocus value. The dependence of the precision on the signal-to-noise ratio will be discussed further on.

Next, we can look into the numerical values that can ultimately be obtained for the precision with which the thickness can be measured. Fig. 4(a) and (b) shows the standard deviation as a function of thickness under the optimal microscope settings for Au[100] and Si[100] atom columns, respectively. For both types of columns, the standard deviations are very low compared to the interatomic distances. This means that the thickness, or, equivalently, the number of atoms in an atom column, can, in principle, be estimated with atomic resolution from electron microscopy images. However, in the interpretation of these numbers, one should keep in mind that the actual precision that can be obtained critically depends on the electron dose. Fig. 5 shows the lower

![Graph](image1.png)

**Fig. 3.** The standard deviation of the thickness estimate as a function of the number of images taken in a focal series for an isolated Au[100] atom column. The thickness corresponds to half an extinction distance • and to one extinction distance ▲. The starting focus is at 50 Å with a defocus step of 50 Å.

![Graph](image2.png)

**Fig. 4.** The standard deviation of the thickness estimate as a function of thickness for isolated (a) Au[100] and (b) Si[100] atom columns using the microscope settings in Tables 1 and 3.
bound on the standard deviation of the thickness estimate as a function of the number of electrons. From Eqs. (19) and (21), it follows that the standard deviation increases by a factor of $\sqrt{C}$ when the number of electrons is reduced by a factor of $C$.

Although the previous example suggests that the thickness of atom columns with one atom difference should be discernable, this is often not the case in practice. There are only few examples in which the number of atoms can be counted up to a limited thickness [33,34]. As explained above, one reason for this discrepancy is the electron dose. Another explanation is absorption. Absorption results from the inelastic interaction between the incident electron beam and the atoms. This includes phonon scattering, plasmon scattering and single electron excitations from inner atom shells [35–37]. Of all these scattering processes, phonon scattering is believed to be the strongest since the cross-section is peaked at the atom core [38]. In order to study this effect, we included absorption in the expectation model by describing the potential as a complex function [39]. The most commonly used assumption for the potential is to add an imaginary part which is proportional to the real potential [37],

$$V = V_R + iV_I = V_R + i\alpha V_R,$$

where $\alpha$ represents the proportionality constant. In [8], it is shown how this extra term in the potential affects the exit wave. The main effect is an exponential decrease of the amplitude which increases with $\alpha$ and the thickness. We use realistic values for $\alpha$ of 0.1, 0.3 and 0.5 in this study. In Fig. 6(a) and (b), results for Au[100] and Si[100] atom columns, respectively, are given for the model including the absorption. The horizontal dashed line represents the maximum allowable standard deviation of the thickness such that two 95% tolerance intervals corresponding to columns with a difference of one atom in a column are just not overlapping. The vertical dashed line indicates the thickness at one extinction distance. From Fig. 6, it is clear that the standard deviation increases with the absorption factor $\alpha$ and with thickness. If $\alpha$ is smaller than 0.3, columns with a difference of one atom are expected to be discernable up to a thickness of one extinction distance for Au[100]. For Si[100], this thickness limit is about two-thirds of the extinction distance. At a given thickness, the precision that can be obtained is dominated by the absorption effect rather than by the particular choice of microscope settings.

So far, the defocus of the image has been assumed to be known a priori. However, on the atomic scale, the defocus value for every atom column is different due to an uneven surface of the object and needs to be estimated along with the other unknown object parameters. Therefore, we studied the effect of including the defocus as an extra unknown parameter. In that case, the vector of parameter values becomes

$$\mathbf{h} = (\beta_x, \beta_y, z, \epsilon).$$

A logical consequence of the larger parameter vector is that the standard deviation of the thickness will increase. The results of this analysis are shown in Fig. 7. Compared to the numerical results in Fig. 6, it can be seen that the standard deviation...
deviation of the thickness estimates increases with a factor of 1.3 on average, for both Au[100] and Si[100] atom columns within one extinction distance. The standard deviation of the defocus estimates as a function of thickness is given in Fig. 8 for Au[100] and Si[100] atom columns. As in Figs. 6 and 7 the horizontal dashed line represents the maximum allowable standard deviation of the thickness such that two 95% tolerance intervals corresponding to columns with a difference of one atom in a column are just not overlapping. The vertical dashed line indicates a thickness of one extinction distance.

4.3.2. Neighboring columns

It was shown in [40] that neighboring columns complicate the visual interpretation of reconstructed exit waves from HRTEM images in terms of the atomic structure. Therefore, it is important to evaluate the effect of neighboring columns on the precision with which the thickness can be estimated. For simplicity, we considered neighboring atom columns of the same type and of the same thickness as the central atom column. The intercolumn distances $d_{A-A}$ we use are given in Table 2. As in the analysis for an isolated atom column, the microscope settings have been optimized. It turns out that they coincide with those given in

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**Fig. 7.** The standard deviation of the thickness estimate as a function of thickness for isolated (a) Au[100] and (b) Si[100] atom columns adding the defocus $\epsilon$ as an extra unknown parameter in $\theta$. The absorption of the atoms is included. The horizontal dashed line represents the maximum allowable standard deviation of the thickness such that two 95% tolerance intervals corresponding to columns with a difference of one atom in a column are just not overlapping. The vertical dashed line indicates a thickness of one extinction distance.

**Fig. 8.** The standard deviation of the defocus estimate as a function of thickness for isolated (a) Au[100] and (b) Si[100] atom columns including the absorption of the atoms. The horizontal dashed line represents the maximum allowable standard deviation of the defocus such that two 95% tolerance intervals corresponding to columns with a surface step of one atom distance are just not overlapping. The vertical dashed line indicates a thickness of one extinction distance.
Table 3. In the case of Au[100], we simulate a $3 \times 3$ lattice of atom columns whereas in the case of Si[100], we simulate a $5 \times 5$ lattice of atom columns. We centered the FOV is at the central atom column. The parameter vector $\theta$ now contains the three-dimensional coordinates of 9 atoms for the Au[100] lattice and 25 atoms for the Si[100] lattice. The effect of neighboring columns on the precision with which the thickness and surface roughness of the central column can be estimated will now be discussed.

First, it is assumed that the defocus of the atom columns is known. In that case, the increase of the standard deviation of the thickness estimate is marginal compared to the case of an isolated atom column. Second, we considered the defocus as an unknown parameter. The defocus value may be different for every atom column since the vertical positions can indeed be different. Therefore, we add an extra parameter to the model for every atom column. This causes an additional increase of the standard deviation. Again, this increase is only marginal when compared to the results given in Fig. 7. In addition, the standard deviation of the defocus estimate hardly differs from the results given in Fig. 8. As a result, the effect of neighboring columns on the precision is only marginal if the intercolumn distance is wide enough. Fig. 9 shows, the relationship between the standard deviation of the thickness estimate and the number of neighboring columns as a function of the intercolumn distance for Au[100] atom columns at half an extinction distance. It shows that the standard deviation increases with decreasing intercolumn distance. Moreover, for small intercolumn distances, the standard deviation of the thickness estimate increases as well with increasing number of neighboring columns. This confirms the expectation that, as the intercolumn distance decreases, the effect of neighboring columns increases, leading to a reduction of contrast and a corresponding reduction of the precision.

4.4. Discussions

In the previous sections, the precision with which the thickness and the defocus can be estimated has been evaluated and optimized as a function of the microscope settings. The precision that can be achieved for a single atom column is affected to a larger extent by absorption due to phonon scattering than by the microscope settings and the influence of neighboring columns. In addition, the actual precision that can be obtained critically depends on the total electron dose. An increase of the electron dose by a factor of $C$ improves the precision by a factor of $\sqrt{C}$. An increase or decrease of the electron dose also affects the critical thickness limit up to which reliable structure information at the atomic scale can be obtained. If we take the proportionality constant of the imaginary potential $\varepsilon = 0.3$, a column thickness smaller than the extinction distance is required in order to make it possible to distinguish between two columns that differ in one atom. Moreover, a column thickness smaller than half the extinction distance is required to detect surface steps of one atom.

The reason why the precision is mostly affected by phonon scattering can directly be related to the reduction of image contrast in the presence of absorption. Reduction of contrast indeed results in a loss of precision [41]. In addition to absorption, there are also other factors that determine the contrast. At present, the contrast mechanism in HRTEM imaging is not yet fully understood. The contrast in experimental images is typically a factor of 3 lower than in simulated images. This is the well-known contrast problem, often referred to as the Snobbs factor [42–44]. Besides phonon scattering, the stability of the specimen and the microscope, atom thermal vibration, lattice configuration, residual microscope aberrations and time-dependent incoherence may also contribute to a loss of image contrast [45].

5. Conclusions

In this paper, we have investigated theoretically to what extent quantitative measurements of the mass, in terms of thickness, and surface profile (defocus) of an atom column, can be obtained from HRTEM images. For this purpose, the statistical precision with which these object parameters can be estimated has been quantified. In order to explore the ultimate limits, the precision has been optimized as a function of the microscope settings. However, it turns out that the precision with which the thickness and surface profile can be estimated is affected by the microscope settings only to a small extent. The main driving factor is the absorption caused by phonon scattering, which reduces the image contrast. Moreover, the precision...
with which the thickness and surface profile can be estimated is poor when compared to the precision with which the atom column positions can be estimated. This indicates that acquiring three-dimensional structure information in terms of thickness or mass is much more difficult than obtaining two-dimensional information on the projected atom column positions. The precision critically depends on the electron dose determining the signal-to-noise ratio. The most precise estimates for the thickness are obtained when all electron counts are used in one single image taken at optimal microscope settings rather than when the electron dose is spread over a focal series of images. Under realistic dose conditions, we found that a column thickness smaller than the extinction distance is required to estimate the mass with single atom sensitivity. Moreover, a column thickness below half the extinction distance is required to detect surface steps of one atom.

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Appendix

If the thickness is estimated using the maximum likelihood estimator, the estimator $\hat{z}$ can be assumed to be normally distributed about the true thickness $z$ with a variance corresponding to the CRLB, $\sigma_z^2$, under the condition that the asymptotic properties of the maximum likelihood estimator hold [1]. This has been tested by means of simulations: 100 different HRTEM experiments made on an isolated Au[100] atom column are simulated. The observations are modeled by means of the parameters given in Table 1 with the optimal conditions for $C$, $C_n$ and $\epsilon$ as given in Table 3. Next, the position coordinates and the thickness of the atom column are estimated from each simulation experiment using the maximum likelihood estimator. Finally, the variance of the thickness estimates is computed and compared with the CRLB on the variance. The CRLB on the variance is computed by substituting the true values of the parameters into Eq. (21). The results are presented in Table 4.

Table 4

<table>
<thead>
<tr>
<th>Column type</th>
<th>CRLB ($\lambda^2$)</th>
<th>Estimated variance ($\lambda^2$)</th>
<th>95% confidence interval for the variance ($\lambda^2$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Isolated column</td>
<td>0.0079</td>
<td>0.0064</td>
<td>[0.0051, 0.0082]</td>
</tr>
<tr>
<td>With neighboring columns</td>
<td>0.0078</td>
<td>0.0096</td>
<td>[0.0077, 0.0123]</td>
</tr>
</tbody>
</table>

References


[45] D. Van Dyck, Strong evidence for phonon scattering as the main cause for Stobbs factor, Ultramicroscopy, submitted for publication.