Refinement of the 200 structure factor for GaAs using parallel and convergent beam electron nanodiffraction data

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We present a new method to measure structure factors from electron spot diffraction patterns recorded under almost parallel illumination in transmission electron microscopes. Bloch wave refinement routines have been developed to refine the crystal thickness, its orientation and structure factors by comparison of experimentally recorded and calculated intensities. Our method requires a modicum of computational effort, making it suitable for contemporary personal computers. Frozen lattice and Bloch wave simulations of GaAs diffraction patterns are used to derive optimised experimental conditions. Systematic errors are estimated from the application of the method to simulated diffraction patterns and rules for the recognition of physically reasonable initial refinement conditions are derived. The method is applied to the measurement of the 200 structure factor for GaAs. We found that the influence of inelastically scattered electrons is negligible. Additionally, we measured the 200 structure factor from zero loss filtered two-dimensional convergent beam electron diffraction patterns. The precision of both methods is found to be comparable and the results agree well with each other. A deviation of more than 20% from isolated atom scattering data is observed, whereas close agreement is found with structure factors obtained from density functional theory [A. Rosenauer, M. Schowalter, F. Glas, D. Lamoen, Phys. Rev. B 72 (2005), 085326-1], which account for the redistribution of electrons due to chemical bonding via modified atomic scattering amplitudes.

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1. Introduction

Evaluations using modern measurement techniques in the field of transmission electron microscopy (TEM) involve simulations as an essential part. The reliability of computer simulations may be seen to be determined by at least three criteria. First, appropriate models are needed to describe the propagation of electrons through the specimen. Bloch wave or multislice methods have proven to be applicable tools in this respect. Second, general parameters such as the thickness, the orientation and the temperature of the specimen should either be measured or their influence should be minimised by finding optimised imaging conditions. Third, any present simulation method relies on atomic scattering amplitudes (ASA) that are mostly obtained from numerical calculations for isolated atoms as published in, e.g., [2–4]. Even from a heuristic point of view it is obvious that treating the electrostatic potential in solids as built up by the potentials of isolated atoms is questionable because of a redistribution of electrons due to chemical bonding. Since chemical bonding mainly affects the outermost orbitals of an atom, structure factors of low order are modified most significantly.

TEM methods for the measurement of low order structure factors traditionally involve the refinement of convergent beam electron diffraction (CBED) patterns [5–7]. However, quantitative CBED requires not only advanced experimental facilities such as energy filters, but also time intensive simulations for the refinement procedures if two-dimensional intensity distributions are to be simulated. For the refinement of crystal structures, these circumstances have motivated the development of the parallel beam electron diffraction (PBED) technique and the program package ELSTRU by Jansen et al. [8]. In the PBED case, the recorded Bragg intensities can be calculated by only one simulation per refinement cycle, so that a modern PC is usually sufficient. Furthermore, Jansen et al. [8] have shown that, under certain circumstances, energy filtering is not necessary for PBED structure refinement [9].

In this article, we present a new method to measure structure factors from PBED patterns. To allow a consolidated discussion of
the results, our studies are concentrated on GaAs, which has been investigated by several groups (e.g. [7,10,11]) in the past. Furthermore, our report is divided into four main sections. In Section 3, the conceptional basics of our Bloch wave refinement routines used by the PBED method are described briefly, followed by a detailed analysis of error sources. This is done via the application of the method to simulated diffraction patterns containing thermal diffuse background, where all parameters of interest are known. Section 4 deals with the measurement of the 200 structure factor for GaAs from experimental PBED patterns. Additionally, we performed CBED measurements of the 200 structure factor which are summarised in Section 5. Section 6 contains a comparison of the PBED and the CBED method and a discussion of the results.

In recent calculations, Rosenauer et al. [1] used density functional theory (DFT) methods to account for bonding effects in ternary sphalerite-type semiconductor materials such as In_{x}Ga_{1−x}As [1,12]. The authors propose a model which allows to retain the structure factor as a central scattering property of a crystal if modified atomic scattering amplitudes (MASA) are used and they give theoretical results for the chemically sensitive 200 crystal. The application of the method to simulated diffraction patterns along incidence directions with permuted and/or inverted particular indices may look nearly identical. For this reason, a CBED pattern was recorded at a thicker specimen region in inverted particular indices may look nearly identical. For this reason, a CBED pattern was recorded at a thicker specimen region containing thermal diffuse background, where all parameters of interest are known. Section 4 deals with the measurement of the 200 structure factor for GaAs from experimental PBED patterns. Additionally, we performed CBED measurements of the 200 structure factor which are summarised in Section 5. Section 6 contains a comparison of the PBED and the CBED method and a discussion of the results.

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Given the technical relevance and the variety of sphalerite-type semiconductor alloys, PBED can serve as an efficient method to verify, e.g., the MASA concept also for crystals which have not yet been subject to detailed studies.

2. Experimental

The specimen used for both the PBED and the CBED work was cut from a standard GaAs wafer in such a way that the surface normal of the specimen and one of the (010) directions include an angle of 31°. After mechanically grinding the specimen by the tripod method, an Ar⁺ ion polishing facility (Gatan) was used for final etching with an ion energy of 3.6 keV and incident angles between 3° and 5°. All diffraction patterns have been recorded at a Titan 80/300 facility operated at 300 kV.

2.1. PBED pattern acquisition

Eleven unfiltered PBED patterns have been recorded on Imaging plates (Ditabis) with exposure times in the range of 5−10 s. Six zero loss energy filtered PBED patterns have also been recorded with a Gatan Imaging Filter (GIF Tridiem 863) on a 2 K × 2 K CCD camera. A slit width of 10 eV and exposure times between 1 and 5 s were used. Among all exposures, thickness and orientation were varied slightly by different beam shift and specimen tilt settings near the zone axis [0 5 3]. The condenser and diffraction lens settings to approach parallel illumination conditions are described in Section 3.2. GaAs is a polar crystal, so neither the preparation nor spot diffraction patterns alone can assure that the intended zone axis is definitely [0 5 3]. Since PBED patterns along incidence directions with permuted and/or inverted particular indices may look nearly identical. For this reason, a CBED pattern was recorded at a thicker specimen region with one of the 200 reflections in Bragg condition.

3. The PBED method

Central features of the PBED method are

(i) the acquisition of diffraction patterns from nanometre-sized areas using a parallel beam,
(ii) the extraction of integrated Bragg intensities and the subtraction of background,
(iii) the estimation of reliable initial thicknesses for the refinement,
(iv) and the Bloch wave simulation of PBED patterns, as well as the least-squares refinement of desired properties.

The necessity of nanodiffraction can be understood from the fact that thickness and orientation vary locally at real specimens. In this respect, we profit from experiences collected with the program MSLS [8] where it was found that probe diameters around 10−15 nm are well suited for quantitative refinements. As too small probes will lead to artefacts in the shape of the Bragg peaks, a lower limit for the probe diameter is derived in this section. Furthermore, the influence of errors induced by a slight beam convergence and the background subtraction is examined quantitatively and rules for the identification of physically plausible initial guesses are deduced.

3.1. The Bloch wave refinement routine

In high energy approximation, the computational kernel contains the diagonalisation of the matrix \( \mathbf{A} \) whose elements are defined by

\[
A_{gh} = U_{g, h} + \delta_{gh}(-2k_0h - h^2).
\]  

In Eq. (1), \( g \) and \( h \) are reciprocal lattice vectors, \( \delta_{gh} \) is the Kronecker symbol. The main diagonal of \( \mathbf{A} \) contains the orientation of the crystal relative to incident electrons via the wave vector \( k_0 \) of the primary wave in the crystal. The full dynamical interaction of all beams which are taken into account in the simulation is carried by the entries \( U_{g, h} \), which
include relativistic effects and the Debye–Waller factors \(\exp(-4\pi^2 (u_t^2)/|\mathbf{g}|^2)\), \(|\mathbf{g}|^2\) being the mean-square displacement of atomic species \(i\).

Results will be given also without relativistic correction to make them independent on the acceleration voltage, whereas the atomic displacement parameters are inherent quantities of the measurements and will be given in terms of the Debye parameter \(B_i = 8\pi^2 (u_t^2)\). In the following, \(U_g\) is referred to as the crystal potential, which is proportional to the structure factor \(F_g\) and has the unit \(1/(\text{length})^2\). We will use the term 200 potential substitutionally for all symmetry related potentials, which are treated as constraint in the refinements. To compare with the results of X-ray studies, our results will also be converted to X-raystructure factors \(\tilde{F}_g^0\) according to [16]

\[
\tilde{F}_g^0 = \sum_{i} Z_i e^{-8\pi^2 |\mathbf{g}|^2/\lambda^2} \exp(-2\pi i \mathbf{g} \cdot \mathbf{r}_i) \frac{2\pi^2 c^2 \Omega |\mathbf{g}|^2 c^2}{E_m e^2} U_g.
\]

Eq. (2) contains Planck’s constant \(\hbar\), the charge \(e\) of an electron and the unit cell volume \(V\). \(E_m\) is the total energy of the incident electrons, \(c\) is the velocity of light in vacuum and \(i\) denotes all atoms in a unit cell at positions \(\mathbf{r}_i\) with atomic numbers \(Z_i\).

The selection of beams follows [17], except that no Bethe approximation is made to reduce the size of \(A\). Because the incidence directions considered here correspond to very sparse zones. The convergence of each refinement result is checked with respect to the inclusion of more beams. Note that the matrix \(A\) shows no symmetries in the general case of noncentro-symmetric crystals with absorption. Throughout this paper, the term absorption refers to losses of electrons in the Bragg scattered beams to the diffuse background in the diffraction pattern caused by thermal diffuse scattering (TDS) processes [2,3].

Diffraction beam amplitudes, phases and intensities are computed from the scattering matrix \(S\) defined by [15]

\[
S = A \times (i.e)^{-1}.
\]

Here, the matrix \(A\) contains all eigenvectors of \(\tilde{A}\). The matrix \(i.e\) is diagonal and introduces the specimen thickness \(t\) and the orientation of the surface normal to the simulation. Its entries are given by \(i.e = \exp(2\pi i \mathbf{g} \cdot \mathbf{n})\), where the Anpassungen \(\gamma_i\) are calculated from the eigenvalues of \(\tilde{A}\), which are set equal to \(2\gamma_i n_{kj}\). Usually the surface normal \(\mathbf{n}\) is assumed to be parallel to the zone axis or incidence direction. However, this approximation should be verified from case to case.

The Bloch wave method was implemented into Matlab routines. Crystal potentials, thickness and orientation can be refined by iteratively changing the entries in the matrix \(A\) or \(\tilde{A}\), respectively. The program can use ASA from Doyle and Turner [4], Kohl and Weickenmeier [2], Bird and King [3] or MASA from Rosenauer et al. [1]. To account for intensity losses to the TDS background, absorptive form factors are taken from [2] or [3]. Then, the crystal potential Fourier components take the form

\[
U_g = \text{Re}(U_g^e) + i \text{Im}(U_g^e) + i \text{Re}(U_g^a) - \text{Im}(U_g^a),
\]

where the superscripts \(e\) and \(a\) denote the contributions from the electrostatic crystal potential and the imaginary (absorptive) part of the optical potential, respectively. In zone axis \(005\), 200 is the only significant reflection of low order. In the case of reciprocal lattice vectors having all even components, the respective crystal potential in Eq. (4) simplifies to

\[
U_g = \text{Re}(U_g^e) + i \text{Re}(U_g^a) = \text{Re}(U_g) + i \text{Im}(U_g).
\]

The real part of \(U_g\) corresponds to electrostatic and the imaginary part to absorptive contributions. To investigate chemical bonding effects, we thus have to refine only the real part of \(U_{200}\) whereas we take the imaginary part from [2].

During the refinement, the simulated intensities must be compared with Bragg intensities extracted from an experimental diffraction pattern and the difference must be minimised. This optimisation is done by the minimisation of the \(R\)-factor defined by

\[
R = \sum_{m} \left( \frac{|I_m - s \cdot I_0|^2}{\sum_{m} |I_m|^2} \right) \text{ with the simplex algorithm.} \]

The superscripts \(E\) and \(S\) indicate experimental and simulated intensities, respectively. The scaling factor \(s\) is used to scale the simulated intensities with respect to the number of counts recorded in the experiment. It is thus equal to the ratio of all summed experimental and all summed simulated intensities and can be adjusted automatically after every refinement cycle. Note that the summation in Eq. (6) usually includes only those reflections which are significantly sensitive to the quantities to be refined. Please also note the similarity to the MSLS principle [8] which uses the same definition of \(R\) as in Eq. (6).

3.2. Practical considerations

Principally, the alignment of the condenser system to approach parallel illumination is straightforward: first, the diffraction lens should be focused on the objective aperture, and second the Bragg spots should be focused by changing the condenser lens excitation, usually by means of the C2 lens. If the objective aperture is not exactly in the focal plane of the objective lens, a slight beam convergence will not be noticed because it is compensated by an overfocus of the diffraction lens. We therefore applied a small beam convergence of 0.5 mrad to the simulation of test patterns (see Section 3.3). Additionally, the realisation of parallel illumination becomes more complicated if probes with diameters below 10–15 nm are required. On the one hand, smaller probes reduce the errors arising from locally varying specimen orientation and thickness gradients. On the other hand, plane wave incidence contradicts nanodiffraction experiments by definition. Note that selected area diffraction (SAD) can be critical due to lens errors occurring in the intermediate image. For this reason the PBED pattern should rather be formed by small probes than by SAD.

To estimate the effect of the Bessel-like broadening of the Bragg spots with decreasing probe diameter, we performed standard Bloch wave simulations of the object exit wave for GaAs crystals in [0 1 0] orientation. The effect of a finite probe diameter has been approached by setting the exit wave function to zero outside a circular region as shown in Fig. 1a, where a probe diameter of 3.5 nm was used. This treatment relies on the fact that we consider only thin specimens with thicknesses of a few tens of nanometres, where the broadening of the beam inside the crystal may be negligible, so that the exit wave function will be nonzero only in a region comparably large as the probe size.

The corresponding diffraction pattern is shown in Fig. 1b in logarithmic scale. Obviously the Bragg intensities spread significantly around their Bragg position, which can lead to problems if Bragg spots and TDS background are to be separated. For GaAs crystals at 300 kV we found that the extension of the Bragg peaks is negligible for spot diameters above 5 nm.

From this we conclude that probe diameters between 5 and 15 nm are well suited for quantitative PBED, whereas the upper limit depends on the homogeneity of specimen thickness and orientation in the illuminated region. This might be checked from case to case by looking at bending and thickness contours in imaging mode. To come to the required probe diameter range, we cut C2 apertures from 3 µm thick platinum foils using a focused ion beam facility. In a FEI Titan 80/300 we achieve parallel probes
being 12 ± 1.5 nm in diameter with a C2 aperture diameter of 8 µm. Further probe size measurements were carried out at a Philips CM 200 with a C2 aperture diameter of 5 µm, leading to a probe size of 7 ± 1.5 nm, which approaches the lower limit estimated above.

### 3.3. Estimation of systematic errors

The PBED method profits from the assumption of plane wave incidence where the illumination can be described by only one primary wave vector \( \mathbf{k}_0 \). A realistic probe in a TEM aligned for parallel illumination contains the effect of the spherical aberration of the condenser lens system, the cutoff of high spatial frequencies due to the micron-sized C2 apertures and a slight misalignment of the diffraction lens focus which leads to a slightly convergent beam, as described in the last subsection. This probe must then be propagated through the specimen by a multislice algorithm to take into account the displacement of the atoms due to their thermal movement. We applied the frozen lattice (FL) approximation where this displacement is uncorrelated among the atoms, yielding the TDS background in between the Bragg reflections together with features like Kikuchi bands. Scattering factors were taken from [2].

The conditions for the FL simulation were similar to those of the actual measurements in Section 4. For a FEI Titan 80/300 operated at 300 kV, a probe was simulated with a \( C \) value of 2 mm for the condenser lens system and a beam convergence of 0.5 mrad was applied, leading to a probe with a full width at half maximum of 10 nm. We simulated three diffraction patterns for GaAs in the zone axis [0 5 3] with thicknesses of 13.19, 23.08 and 32.96 nm, respectively. Debye parameters of \( B_{Ga} = 0.7 \text{ Å}^2 \) and \( B_{As} = 0.6 \text{ Å}^2 \) were chosen which correspond to a temperature of 300 K [18]. The lateral extension of the supercell was 40 × 40 nm².

A part of the 32.96 nm simulation is shown in Fig. 2 with emphasis on the TDS background. ZOLZ reflections are encircled and form the experimental data set for the minimisation of \( R \) in Eq. (6). The respective Bragg intensities were extracted using the program GREED, which is part of the ELSTRU software package as MSLS [8]. Main steps of the GREED routine are the identification and indexing of the peaks, followed by the data reduction which subtracts the local background. We excluded the primary beam from this procedure because its intensity is usually not available in the experiment due to a saturation of the recording medium. An inset in Fig. 2 depicts the probe that was propagated through the sample to illustrate that the illumination is not homogeneous throughout the specimen surface. Besides the encircled ZOLZ peaks, HOLZ reflections are present as shown in Fig. 2, so that HOLZ are included in all Bloch wave simulations.

We computed the \( R \)-value as a function of the specimen thickness (thickness scan) for each data set to find initial thickness guesses for the refinement. The curves \( R(t) \) are shown in Fig. 3. In this case we used already the correct zone axis orientation and also the scattering data from [2]. From Eq. (6) we therefore expect the \( R \)-value to drop to zero for the thicknesses used in the FL simulations if the background subtraction was exact. For each of the three thickness scans, we see that this is indeed approximately the case, even if there are several comparably deep minima for the 32.96 nm simulation, a point addressed later.

With an initial thickness derived from the thickness scan, we started the refinement of the thickness, the centre of Laue circle (COLC) and the real part of the 200 potential. This was done to study how accurately the quantities which are of interest in real
measurements are represented at the minimum of the $R$-value. Starting thicknesses, refined quantities and the minimised $R$-values are listed in Table 1. From the $R$-values we see that systematic errors arising from the background subtraction and from the assumption of plane wave incidence raise the $R$-value less than 1% in all three cases. To judge the quality of the refinements, one has to keep in mind that the optimisation takes place in a multidimensional parameter space, where several constellations of parameters may lead to the same $R$-value. In our case this space is five dimensional if we include the scaling factor in the refinement, so that the resulting thicknesses, COLC and the 200 crystal potential were simultaneously refined for each data set separately. The last line lists the optimised $R$-value.

Table 1

Refinement results for the three simulated data sets.

<table>
<thead>
<tr>
<th>Thickness [nm]</th>
<th>13.19 nm</th>
<th>23.08 nm</th>
<th>32.96 nm</th>
</tr>
</thead>
<tbody>
<tr>
<td>$t_0$ (nm)</td>
<td>13.07</td>
<td>23.16</td>
<td>33.01</td>
</tr>
<tr>
<td>$t$ (nm)</td>
<td>13.07</td>
<td>23.24</td>
<td>33.09</td>
</tr>
<tr>
<td>COLC</td>
<td>(000)</td>
<td>(000)</td>
<td>(000)</td>
</tr>
<tr>
<td>$\text{Re}(U_{200})$ (nm$^{-2}$)</td>
<td>-0.576</td>
<td>-0.572</td>
<td>-0.556</td>
</tr>
<tr>
<td>$R$</td>
<td>0.1%</td>
<td>0.8%</td>
<td>0.20%</td>
</tr>
</tbody>
</table>

Column titles refer to the true thicknesses used in the FL simulation. The refinements started with the thicknesses $t_0$. Thickness, COLC and the 200 crystal potential were simultaneously refined for each data set separately. The last line lists the optimised $R$-value.

3.4. Ambiguity of refinement results

In practice, the situation is different from that considered in Section 3.3 because the refinement aims at determining low order structure factors whose values may be needed already for the thickness scan. Excluding the respective reflections from the thickness scan has proven to give the best results in this case. However, the interaction of all scattered beams will still lead to inaccurate starting values for the thickness. For example, the deepness of the minima shown in Fig. 3 can depend on the magnitude of low order structure factors, so that the global minimum need not be the true one. To simulate this case, we repeated the thickness scans from Fig. 3 with $\text{Re}(U_{200}) = -0.666$ nm$^{-2}$ and excluded the 200 intensities from the computation of the $R$-value and the scaling factor. Thus we simulate the situation where the experimental data set corresponds to a crystal potential $U_{200}$ that is 0.12 nm$^{-2}$ larger than the potential used for the thickness scan. The resulting $R(t)$ curves are depicted in Fig. 4. It can clearly be seen that several minima of comparable deepness occur because the information of the 200 beams is missing. But it also becomes obvious that there is a minimum close to the true position in every case. In experiments, any of these minima may be the true one if the thickness is not measured separately by different methods. In the following, we will discuss the plausibility of each of these possible initial thickness guesses and give rules to identify the physically reliable one.

It is in the nature of refinement purposes that the system is already well understood, so that only some parameters will be modified slightly. In our case, these parameters are the low order crystal potentials which are modified due to chemical bonding. However, the scattering factors from Kohl and Weickenmeier [2] are assumed to be valid for higher momentum transfers. Therefore, the plausibility of refinement results or initial guesses can be checked by verifying that known quantities are reproduced. As an example, we will consider the possible initial guesses for the specimen thickness of the 32.96 nm simulation. From Fig. 4, seven possible starting thicknesses $t_0$ were derived from the $R$-value minima positions. For each of these thicknesses, we calculated the $R$-value as a function of $\text{Re}(U_{400})$ in a range $\text{Re}(U_{400}) \in [3-6]$ nm$^{-2}$. In these $R(U)$ curves, the $R$-value was computed from the 400 intensities only and is plotted in Fig. 5 in logarithmic scale. The true real part of $U_{400}$ which was used in the FL simulations is marked by the black triangle.

If the correct thickness and 200 potential entered the simulation of the $R(U)$ curves shown in Fig. 5, we would see a minimum at the position of the black triangle. But as the
Best agreement with the values used in the FL simulation within the errors we derived in Section 3.3.

Few was said as to the refinement of the orientation. This is because the results were always appreciably accurate when the initial guess for the COLC did not deviate more than 0.5 reciprocal basis vectors from the true one, an accuracy which can easily be achieved by evaluating the intersections of Kikuchi bands.

We close our studies of ambiguous refinement results with a comment concerning the astonishing similarity between the thickness scans of the 13.19 nm and the 32.96 nm simulations shown in Fig. 4. Both thicknesses fall into a region where the 400 reflections have intensity maxima and strongly dominate the diffraction pattern and thus the denominator of Eq. (6). The \( R \)-value can in both cases only rise significantly at those thicknesses where 400 is weak (which is the case near 20 and 40 nm), because only then the differences in the nominator of Eq. (6) can become comparably large as the denominator. Critically examining the refinement results for unknown properties by checking the outcome for known properties is thus an essential part of the PBED method. For the 13.19 nm simulation, the 200, 400 and 800 potentials are refined to their true values only if a starting thickness around 13 nm is used.

### 4. Measurement of crystal potentials for GaAs using the PBED method

#### 4.1. Estimation of initial thicknesses for the refinement

For every data set, an initial COLC was estimated by evaluating the intersections of Kikuchi bands. In order to find initial guesses for the thickness refinement, we calculated \( R(t) \) curves for each set as done in Section 3.4. To show that the evaluation of real PBED patterns is strictly analogous to that of the simulated ones, we exemplify the procedure using only one experimental pattern for reasons of brevity.

A typical thickness scan is shown in Fig. 6 for one data set (set No. 1 in Table 3) where no energy filter was used. Note that the 200 intensities have been excluded from the computation of the \( R \)-value and the scaling factor. The function \( R(t) \) exhibits six local minima with \( R \)-values below 5\%, whose positions mark possible starting thicknesses for the refinement. To identify the
most plausible thickness, six $R(U_{200})$ curves are depicted in Fig. 7 which correspond to the thicknesses taken at the minima in Fig. 6. The curves for $t_0 = 14.57$, 38.94 and 43.47 nm have no minimum at all near the potential we expect from [2], which is indicated by the black triangle. Thicknesses $t_0 = 3.51$ and 33.42 nm show minima around $R(U_{200}) = 3.5$ nm$^{-2}$, but the curve corresponding to $t_0 = 24.12$ nm is much more plausible, not only because its minimum $R$-value is nearly an order of magnitude lower, but also because the minimum position is closest to what we expect from the isolated atom approximation. In this way, we found initial thicknesses for 6 of the 11 unfiltered and for 3 of the 6 zero loss filtered data sets. For the remaining sets, the $R$-values either exceeded 30% at the minima of $R(U_{200})$ or no minimum did show up at all in a physically plausible range. Hence nine sets are left for the refinement.

### 4.2. Refinement results for the 200 potential

The final refinement results were obtained from mainly three subsequent steps. As the specimen thickness and the orientation are considered to contain the major errors in the beginning, both quantities were refined simultaneously in a first run without using the 200 intensities. Second, solely the 200 potential was refined using the thickness and COLC results from the first step. The refinement starting value $R(U_{200}) = -0.546$ nm$^{-2}$ was calculated according to [2]. Third, all three parameters thickness, COLC and $R(U_{200})$ were refined simultaneously using the results from the first two steps as input. The scaling factor was always included in the refinements and each data set was treated separately. Typically, convergence was achieved after approximately 20 iterations in the last refinement step, which took roughly 5–10 min on a 2.66 GHz PC. Depending on the orientation, 780–800 beams from four Laue zones have been included in the calculation. Debye parameters of $B_{as} = 0.68$ Å$^2$ and $B_{as} = 0.59$ Å$^2$ were used which correspond to a temperature of 295 K according to [18]. The assumption that the specimen is at room temperature is based on the fact that our 8 μm C2 aperture drastically decreases the beam current density. The refinement results are shown in Table 3.

From the initial and refined thicknesses shown in Table 3, it can be seen that our preliminary thickness guesses $t_0$ are accurate to a nanometre for all data sets. Furthermore, the COLC reveal that all specimen orientations are close to the zone axis [0 0 3] since the largest tilt of 2.51 mrad occurs for set No. 3. A qualitative comparison of the refinement results for $R(U_{200})$ in the fifth column of Table 3 with the starting value of $R(U_{200}) = -0.546$ nm$^{-2}$ already exhibits a deviation of more than 0.1 nm$^{-2}$ towards larger values for the 200 potential in any of the nine refinements.

As to the effect of zero loss filtering, neither the $R$-values nor the refined potentials of sets Nos. 7–9 indicate that an energy filter is necessary, since they are in good agreement with the results of the unfiltered data sets.

Except for set No. 4, the $R$-values are below 5%, indicating reliable results. Set No. 4 is exceptional also concerning the 200 potential, which is largest here. Nevertheless, we see that $R(U_{200})$ for set No. 9 is nearly comparably large, however, with a very small $R$-value of 2.4%. Since an $R$-value of 7% is still reasonable, set No. 4 has also been included into the statistical evaluation, which is shown at the bottom of Table 3. In comparison to the isolated atom approximation [2,3], the average real part of the 200 potential changed by more than 20%, which is discussed in Section 6.

### 4.3. Behaviour of the 400 crystal potential

In contrast to the 200 potential, the 400 potential is not expected to be significantly modified by chemical bonding. Therefore, we used these potentials for plausibility arguments in Section 4.2 to decide which specimen thickness should enter the refinement. The minimum of the solid curve shown in Fig. 7 is indeed closest to what one expects from the isolated atom approximation, but nevertheless there is no exact coincidence. To judge whether this deviation is in fact due to the wrong 200 potential used for the $R$-value calculation shown in Fig. 7, the behaviour of the curve $R(U_{400})$ before and after the refinement of the 200 potential is depicted in Fig. 8 for each set.

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**Table 3**

Refinement results of the PBED method for nine experimental data sets.

<table>
<thead>
<tr>
<th>No.</th>
<th>$t_0$ (nm)</th>
<th>$t$ (nm)</th>
<th>COLC</th>
<th>$R(U_{200})$ (nm$^{-2}$)</th>
<th>$R$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>24.12</td>
<td>24.07</td>
<td>(0.00 0.00 0.00)</td>
<td>-0.419</td>
<td>0.023</td>
</tr>
<tr>
<td>2</td>
<td>15.52</td>
<td>15.50</td>
<td>(0.96 0.00 0.00)</td>
<td>-0.413</td>
<td>0.040</td>
</tr>
<tr>
<td>3</td>
<td>25.17</td>
<td>25.01</td>
<td>(1.42 0.13 0.72)</td>
<td>-0.435</td>
<td>0.048</td>
</tr>
<tr>
<td>4</td>
<td>23.45</td>
<td>22.59</td>
<td>(0.00 0.00 0.00)</td>
<td>-0.347</td>
<td>0.073</td>
</tr>
<tr>
<td>5</td>
<td>28.45</td>
<td>28.56</td>
<td>(0.41 0.07 0.39)</td>
<td>-0.395</td>
<td>0.027</td>
</tr>
<tr>
<td>6</td>
<td>25.02</td>
<td>24.84</td>
<td>(0.37 0.15 0.24)</td>
<td>-0.393</td>
<td>0.042</td>
</tr>
<tr>
<td>7</td>
<td>20.52</td>
<td>20.32</td>
<td>(0.21 0.00 0.00)</td>
<td>-0.421</td>
<td>0.024</td>
</tr>
<tr>
<td>8</td>
<td>18.72</td>
<td>18.44</td>
<td>(0.38 0.04 0.05)</td>
<td>-0.418</td>
<td>0.041</td>
</tr>
<tr>
<td>9</td>
<td>22.93</td>
<td>22.93</td>
<td>(0.28 0.15 0.27)</td>
<td>-0.361</td>
<td>0.024</td>
</tr>
</tbody>
</table>

Initial and refined thicknesses correspond to the columns $t_0$ and $t$, respectively. The last three columns list the refined COLC and the 200 potential (real part) with the minimised $R$-value, the lower part shows the statistical evaluation.

**Fig. 7.** Dependence of the $R$-value on the 400 crystal potentials (real part) for an experimental PBED pattern. Different curves correspond to different thicknesses $t_0$ (see the legend) taken from the minima positions in Fig. 6. Because the $R$-values cover a wide range, the $R$-axis is scaled logarithmically. The black triangle marks the 400 potential computed from [2].
Fig. 8. Dependence of the R-value on the 400 crystal potential (real part) for the experimental data sets (a) before and (b) after the refinement of the 200 potential. The curves can be assigned to the results in Table 3 with the list in the legend. The black triangle marks the 400 potential computed from [2].

Fig. 8a depicts the results before the refinement of the 200 potential, i.e. only the thickness and the orientations had been optimised using isolated atom scattering data. Obviously, a 400 potential of \( \text{Re}(U_{400}) \approx 5.2 \text{ nm}^{-2} \) is found for most data sets, which is about 0.6 nm\(^{-2} \) larger than the potential according to [2] (black triangle). In contrast to this, Fig. 8b clearly reveals a much better agreement between the R-value minima of each set and the isolated atom approximation. However, even in Fig. 8b the minima scatter significantly around their average position. In this case, this is not severe because the worst R-value is still only 2% (set No. 4) for the isolated atom potential. Furthermore, it is exactly this set which showed the largest deviation of \( \text{Re}(U_{400}) \) during the refinements in Section 4.2. We will not go into further details of each curve shown in Fig. 8, but give average results for \( \text{Re}(U_{400}) \) instead, which we deduced from the positions of the minima. From data sets Nos. 1–9 we obtain \( \text{Re}(U_{400}) = 5.238 \pm 0.284 \text{ nm}^{-2} \) for Fig. 8a and \( \text{Re}(U_{400}) = 4.5042 \pm 0.167 \text{ nm}^{-2} \) for Fig. 8b. Thus the 400 potential for isolated atoms of 4.616 nm\(^{-2} \) only agrees with experiment if our refinement result for \( \text{Re}(U_{200}) \) is used.

5. Measurement of GaAs structure factors using CBED

5.1. Methodical summary

Tsuda and Tanaka [6] have presented a method to refine crystal structural parameters such as atom positions, Debye parameters and low order structure factors from two-dimensional, zero loss filtered CBED patterns. The method will be outlined briefly here (for details, see [6,5]). Their method contains computer programs for the preprocessing of raw experimental data, as well as for the nonlinear least-squares fitting by many beam Bloch wave simulations (program MBFIT). During preprocessing, distortions of the CBED discs and deviations from the intended disc positions due to aberrations of the optical system are corrected for by estimating and refining the elliptical distortion coefficients, as well as the disc shifts. A subtraction of the diffuse background along a given direction (usually parallel to a Kikuchi band) is performed by linear interpolation between the background intensities just outside the disc.

The program MBFIT minimises the factor S defined by

\[
S = \sum_m w_m L_m, \quad \text{GOF} = \sqrt{\frac{S}{N-M}},
\]

where the superscript indicates experimental and simulated intensities, respectively. The scaling factor \( s \) is defined as in Eq. (6). The weights \( w_m \) are derived from the error due to electron counting statistics and the error induced by the background subtraction. Depending on the quantities to be refined, the weights of reflections which are most sensitive to the respective parameter changes may be emphasised manually. Two further quantities, the reliability factor \( R_w \) and the goodness of fit (GOF), defined by

\[
R_w = \frac{S}{\sum_m w_m L_m}, \quad \text{GOF} = \sqrt{\frac{S}{N-M}}
\]

are calculated to measure the quality of the refinement. In Eq. (8), \( N \) and \( M \) denote the number of data points and the number of refined parameters, respectively. The procedure to select and calculate the potentials which are to be included in the calculation is similar to that described in Section 3.1, except that the program can account for further potentials via the generalised-Bethe-potential method [19]. Scattering factors for isolated atoms are used according to [3].

5.2. Refinement preconditions

All CBED patterns have been preprocessed as described in Section 5.1 and [6]. See Section 2.2 for experimental details.

5.2.1. Acceleration voltage of the TEM

The acceleration voltage must be measured separately because the positions of HOLZ lines may depend significantly on it. We therefore simulated GaAs CBED patterns in [0 5 3] zone axis with \( 200 \) in Bragg condition using 16 different voltages between 295 and 305 kV. The ratio \( a/b \) from the intersection of HOLZ lines in the primary disc was determined as shown in Fig. 9. First, straight lines have been marked by eye along the respective HOLZ lines. Second, a number of perpendicular intensity line scans have been calculated, so that the minimum of each line scan yields one position on the HOLZ line. Third, these minimum positions were fitted to obtain four linear equations from which we calculated the
The ratio $a/b$. The actual accelerating voltage was finally obtained by comparing the experimental $a/b$ ratio with the calibration curve, leading to a voltage of 302.1 kV. As HOLZ line positions may also depend on the lattice parameters, and hence on the temperature, we checked the reliability of the result by simulating a CBED pattern for 302.1 kV using lattice parameters corresponding to a temperature of 350 K. The ratio $a/b$ obtained from this pattern still matched the one found above.

5.2.2. Debye parameters

Due to the comparably large C2 aperture and the focusing of the probe, high beam current densities are present in the investigated specimen area, leading to an unknown increase of the temperature and hence the Debye parameters. We used the HOLZ reflections of the CBED pattern recorded with a smaller camera length to refine $B_{Ga}$ and $B_{M}$ and then kept these Debye parameters fixed throughout the structure factor refinements. During the refinement of the Debye parameters, isolated atom scattering data according to [3] were used. The signal of the HOLZ reflections is relatively low because we had to take care that the primary beam would not damage the CCD chip. However, three significant HOLZ reflections could be extracted from the raw data and were used to refine the Debye parameters, starting from the values for room temperature given in Section 4. Before, the thickness and the scaling factor had been determined from the 400 discs. Observed and calculated discs as well as the difference for the refinement result of $B_{Ga} = 0.76 \text{Å}$ and $B_{M} = 0.62 \text{Å}$ are depicted in Table 4. It can be seen that the refinement is more accurate along the center of the respective HOLZ line, whereas the calculation does not contain the blurring of the lines. This may be due to the fact that we did not convolute the calculated intensities with the instrumental function, which is analysed in [6]. Since determining the Debye parameters is not our main goal in this article, we neglect the slight changes introduced by the instrumental function here.

5.2.3. Initial thicknesses

For each data set, most plausible thicknesses and scaling factors have been determined from the GOF plotted versus the specimen thickness (analogous to the $R(t)$ curves in Section 4). Before, the orientations had been found from the deficiency HOLZ lines inside the ZOLZ discs. A typical example for one data set (set No. 1 in Table 5) is depicted in Fig. 10.

Obviously a thickness $t_0 = 145 \text{ nm}$ is a good starting point because this minimum drops to a value $GOF = 6.285$, which is by far the lowest in the plotted thickness range. The fact that the thickness scan leads to an unambiguous result in the CBED case has its origin in the thickness fringes of the 400 disc, which is shown as an inset in Fig. 10. See Table 5 for the starting thicknesses $t_0$ for the other data sets.

5.3. Refinement results for the 200 structure factor

The intensity distribution results for the 400 disc has initially been used to refine the thicknesses and the scaling factors, starting from the results of the thickness scan (Section 5.2.3). Then, the 200 disc was also included in the refinement of the thickness, the scaling factor and $\text{Re}(U_{200})$, the latter starting at $-0.534 \text{ nm}$, which corresponds to the isolated atom data from [3]. Using the resulting thickness and the resulting scaling factor of this refinement, the real part of the 200 potential has been refined to about $0.1 \text{ nm}^{-2}$ larger values, compared to isolated atom data. The HOLZ line patterns show that the orientation varies among different data sets, and so does the thickness $t$.

Although the initial thickness guesses $t_0$ have been stated to be unique according to Fig. 10, the refined thicknesses deviate considerably from the starting values (more than $10 \text{ nm}$ for set No. 2). This may be due to the fact that crystal potentials for isolated atoms had been used during the thickness scans. However, most $R_w$ values do not exceed $7\%$, indicating reliable results. The worst $R_w$ of 9.8% and GOF of 2.486 result for set No. 4, which was nevertheless included in the statistical evaluation.
shown at the bottom of Table 5, because neither the visualisation of the difference between calculation and experiment nor the resulting potential give reason to doubt this result, if we compare with the rest of the table. Comparing the average 200 potential from Table 5 with the isolated atom scattering data [2,3], we realise that the real part changed by ca. 20%. This is similar to the PBED result and will be discussed in the next section.

6. Discussion

6.1. Validity of the PBED test results

Our report on the test of the PBED method has been restricted to GaAs in zone axis [0 5 3] to approach the conditions of the actual measurements. However, the errors induced by, e.g., the background subtraction or the assumption of plane wave incidence can be thought to be extensively independent on the material and the zone axis.

The resulting R-values in Tables 1 and 2 appear to be extremely low, since experimental R-values are a few percent in magnitude (Table 3). This just expresses that slightly convergent aberration-manipulated probes together with the background subtraction are
not responsible for the increase of the minimal R-value in practice. Instead, amorphous layers on the specimen surfaces, variations of the thickness and the orientation throughout the illuminated area and Poissonian noise may govern discrepancies between experimental and simulated data sets. Concerning amorphous layers, the background subtraction will be accurate only if no significant rings form in reciprocal space, as pointed out in [8]. Analogously to this reference, we tested the effect of a thickness gradient in the illuminated area exemplarily for set No. 1 in Table 3, i.e. the experimental data set was compared with the average over five simulated diffraction patterns in the range between 23 and 25 nm. The refinement results \( t = 23.9 \text{ nm} \) and \( \text{Re}(U_{200}) = 0.412 \text{ nm}^{-2} \) show that nearly the average thickness is obtained and that the influence on the refinement of the potential is very low.

Except for the magnitude of the R-values the evaluation of the test patterns in Section 3 and the experimental data sets in Section 4 was analogous. This is an important point because we derived rules for the identification of plausible results, e.g., of the thickness scan using the test data sets and applied the same rules to the experimental situation. Furthermore, the errors of the refinement of the 200 potential are of the same magnitude, indicating plausible test conditions.

### 6.2. Methodological aspects

The results obtained in this work by both the PBED and the CBED method are summarised without relativistic correction in Table 6, which contains also theoretical values expected for isolated atoms and from the MASA concept. Related properties such as the Coulomb potential in \( V \), the structure factor in nm and the X-ray structure factor are also given. At first, our focus is on the measurement to compare PBED and CBED methodologically.

Strictly speaking, the mean results for \( \text{Re}(U_{200}) \) may not be compared directly because of the different Debye parameters used. Nevertheless, the influence of the Debye parameters on the 200 potential is very low because the impact of Gaussian damping decreases with decreasing momentum transfer, and because the difference between the temperatures in both experiments is approximately only 30 K [18]. Hence both the PBED and the CBED method lead to 200 potentials which agree well with each other within the margin of error, which is given in brackets for \( \text{Re}(U_{200}) \) in Table 6. However, knowledge of the Debye parameters is still important to measure the correct specimen thickness, because both quantities may be correlated as to the minimisation of \( R \) in the PBED case [8]. Furthermore, the \( R \)-values of a few percent in Tables 3 and 5 agree well with the \( R \)-values in earlier publications of PBED refinements [8,9] and CBED results obtained with the MBFIT program [6]. The latter reference also contains maximum \( \text{GOF} \) around 2, as in the present work.

The fact that both methods show a different average 200 potential was also investigated with respect to the different scattering data sets [2,3] used. The PBED refinements based on [2,3] yield identical averages and standard deviations.

Regarding the precision of both methods, Table 6 exhibits nearly the same standard deviations. This is surprising since the outcome of the refinement of one CBED data set is in fact already the average over thousands of diffraction patterns, each corresponding to one pixel (and therefore one orientation) in the CBED disc. In our experience, errors for a single refinement derived from standard error propagation rules alone as described in, e.g., [16] are usually unphysically low due to the large number of data points. For this reason, we preferred a statistical evaluation of several independent measurements, resulting in more realistic standard deviations.

The simultaneous refinement of the thickness, the orientation and the 200 crystal potential by the PBED method took a few minutes on one 2.66 GHz CPU, whereas only one refinement cycle for the refinement of the 200 potential alone would have occupied one CPU for nearly 10 h using the CBED method with the same number of beams. CBED therefore requires computer clusters if two-dimensional diffraction patterns are to be simulated.

Care must be taken when comparing the precisions and the calculation times only. Table 5 shows that the refinement results of the CBED method can be judged by visual control of the difference plots. In principle, this is also possible for the Bragg intensities of the PBED patterns, but of course the explanatory power is much less in this case. It is connected with this aspect that the thickness scans look much more unique in the CBED case due to the fringes in the 400 disc, an information which is missing in PBED patterns. Here, only the dynamical interaction of all scattered beams can be used for the thickness estimate. In our experience, this is in principle no limitation because in most cases, the correct starting thickness can be found by checking the behaviour of other quantities which are known with adequate accuracy. In this paper, the 400 potential was used for this purpose. It should be noticed that the thicknesses obtained from our PBED patterns are much smaller than those measured from the CBED patterns, owing to the fact that we tested our refinement results for the thickness and for the orientation with the output of MSLS, which only applies to thicknesses below 15–40 nm [8].

For the zero loss filtered diffraction patterns analysed in this work, it was assumed that energy filtering contributes to the scaling factor only, which is common to all reflections. However, the PBED results show that energy filtering does not alter the refinement results, whereas it is essential for achieving high precision in CBED work [6].

It is worth mentioning that we used the accurately measured acceleration voltage of the TEM for the CBED refinements, but not for the PBED method, where we took the nominal value indicated at the TEM. To estimate the error induced by this approximation, we repeated the refinements from Section 4.2 with the voltage measured by CBED in Section 5.2.1, assuming a deviation of 2.1 kV from 300 kV to be a typical error of the voltage indicated at the TEM. The resulting 200 potential deviated less than 0.003 nm\(^{-2}\) from the results in Table 3 (which correspond to a voltage of 300 kV). Hence the systematic errors are larger than the effect of a slightly modified acceleration voltage.

### Table 6

Collection of PBED and CBED results obtained in this work together with different theoretical data sets according to Weickenmeier and Kohl (WK [2]), Bird and King (BK [3]) and Roseneau et al. (MASA [1]).

| \( \text{Re}(U_{200}) \) (nm\(^{-2}\)) | \( \text{Re}(V_{200}) \) (V) | \( \text{Re}(F_{200}) \) (nm) | \( F_{100}^2 \)
<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>PBED</td>
<td>-0.252 (0.019)</td>
<td>-0.379</td>
<td>-0.143</td>
</tr>
<tr>
<td>WK</td>
<td>-0.346</td>
<td>-0.520</td>
<td>-0.196</td>
</tr>
<tr>
<td>BK</td>
<td>-0.331</td>
<td>-0.498</td>
<td>-0.188</td>
</tr>
<tr>
<td>MASA</td>
<td>-0.268</td>
<td>-0.403</td>
<td>-0.152</td>
</tr>
</tbody>
</table>

**Debye parameters:** \( R_C = 0.68 \text{ \AA}^2 \) and \( B_K = 0.59 \text{ \AA}^2 \)

| \( \text{Re}(U_{200}) \) (nm\(^{-2}\)) | \( \text{Re}(V_{200}) \) (V) | \( \text{Re}(F_{200}) \) (nm) | \( F_{100}^2 \)
<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>CBED</td>
<td>-0.273 (0.018)</td>
<td>-0.411</td>
<td>-0.155</td>
</tr>
<tr>
<td>WK</td>
<td>-0.350</td>
<td>-0.526</td>
<td>-0.199</td>
</tr>
<tr>
<td>BK</td>
<td>-0.335</td>
<td>-0.504</td>
<td>-0.190</td>
</tr>
<tr>
<td>MASA</td>
<td>-0.272</td>
<td>-0.409</td>
<td>-0.154</td>
</tr>
</tbody>
</table>

**Debye parameters:** \( R_C = 0.76 \text{ \AA}^2 \) and \( B_K = 0.62 \text{ \AA}^2 \)

Note that different Debye parameters are involved in PBED and CBED and that all values are nonrelativistic. The standard deviations of the \( \text{Re}(U_{200}) \) refinements are given in brackets and correspond to Tables 5 and 3. Related crystal properties like the structure factor \( F \), the Coulomb potential \( V \) and the X-ray structure factor \( F_{100} \) are also listed.
6.3. Discussion of the results for the 200 potential

Crystal potentials of GaAs have in the past been studied experimentally by several groups with different methods under various experimental conditions. Zuo et al. [7] found \( V_{200} \) to be 0.43 ± 0.008 V from measurements at 90 K and 120 kV by fitting one-dimensional CBED intensity profiles. Since the temperatures of the present work are more than 200 K larger, a direct comparison of the results is hardly possible, because the ratio \( B_{Ga} / B_{As} \) changes significantly from low to high temperatures. This means that the contribution of Ga atoms (relative to that of the As atoms) to the structure factor increases [20,21] \( B_{Ga} < B_{As} \) or decreases [18] \( B_{Ga} > B_{As} \) with increasing temperature. However, referring to the isolated atom approximation [2] and Debye parameters from [20] for 90 K, the result from Zuo et al. [7] corresponds to a change by a factor of 0.83, which deviates a few percent from what we expect from Table 6. As to the error margins given in [7], our measurements seem to be much less precise on the first view, since our standard deviations are larger by a factor of approximately 3.5. We suppose that Zuo et al. [7] found their errors from standard error propagation rules for a single refinement, being much smaller in practice than our statistical errors, as mentioned in Section 6.2.

Stahn et al. [10] measured structure factor amplitudes and Debye parameters by X-ray studies and found \( F_{200}^X \) = 6.34 with an error of 1−2% at 297.65 K. For 287.15 K, they found \( B_{Ga} = 0.666 \AA^2 \) and \( B_{As} = 0.566 \AA^2 \), which agrees well with the data in [18] where we took the Debye parameters for the PBED refinements. Additionally, our CBED measurements confirm that \( B_{Ga} > B_{As} \), unlike [20,21]. Comparing \( F_{200}^X \) from Stahn et al. [10] with the experimental values given in Table 6, all three measurements are in very good agreement. The differences do not exceed 0.026, which is less than ¼ of the error \( \sigma \) of our measurements. Analogous arguments hold for the comparison with the review of Pietsch and Hansen [11] who gave experimental results in the range \( F_{200}^X \in [5.43−6.79] \) with a mean value of 6.10 and a standard deviation of 0.484.

We close this discussion with the comparison of the measured 200 crystal potential with that expected theoretically from the isolated atom approximation [23] and from the MASA concept [1]. As can be seen from Re\(U_{200}\) in Table 6, both the CBED and the PBED measurements agree only with the MASA concept within the margins of error, whereas the isolated atom potentials deviate more than 20% and cannot be verified. Discussing the accuracy of both the PBED and the CBED method would imply that the true 200 potential is known. Granted that the DFT potentials are true, the CBED method would be more accurate, since the result matches the DFT value nearly perfectly. However, one has to keep in mind that seven of the nine PBED results are very close to what is expected from the MASA model, and only measurements Nos. 4 and 9 yield considerably larger potentials, which has significant impact on the statistical average.

Using the PBED method, we have also shown that the 400 potential according to the isolated atom approximation only agrees with the experiment if we use our refined 200 potential for the simulation of diffraction patterns. This additionally confirms the MASA concept which matches with isolated atom scattering data asymptotically for high momentum transfers. For the 400 potential, a difference of 0.3% is expected, which is smaller than the experimental errors.

7. Conclusions

The diffraction of parallel electron nanoprobe can be used to measure the specimen thickness, the specimen orientation and crystal potential Fourier components accurately with a modicum of experimental and computational effort. This makes the method suitable for deducing statistical averages from multiple independent measurements or for the measurement of structure factors for a variety of materials. A C2 aperture of a few microns in diameter in combination with a field emission gun is recommended to obtain nearly parallel probes and significant beam current densities at the same time. Still, the beam current is very low in the PBED case, making specimen heating negligible unlike for the focused probes used in CBED measurements. This means that Debye parameters for the temperature of the specimen holder can be taken from the literature, so that their refinement is not necessary. However, it has been shown already that the PBED method can also be used to refine Debye parameters from ZOLZ reflections [8]. Including HOLZ reflections for this purpose (as in our CBED measurements) is expected to improve the accuracy further, which may be a topic for future work.

A considerable advantage of the CBED method is the possibility to check refinement results visually by comparing the internal structure of the diffraction discs. However, the necessity of an accurate high voltage measurement increases the effort slightly, whereas zero loss energy filtering and the use of computer clusters for simulating whole CBED discs are disadvantages from a financial point of view.

The MASA concept [1] is verified for GaAs by both the PBED and the CBED measurements in this work. This will have important consequences for the CELFA method, if the validity of the MASA can additionally be confirmed for InAs. Then, the accuracy of the measurement of the local concentration in In\(\text{Ga}_{1−x}\)As can be improved drastically since the structure factor for the alloy can be well approximated by linear interpolation [1]. As the CELFA method applies to many other sphalerite-type materials, the PBED method together with the experimental conditions presented here is one possible approach for future measurements of the respective chemically sensitive 200 potentials.

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