Local strain fields in Al-based metal matrix composites

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Local Strain Fields in Al-based Metal Matrix Composites

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Summary

In the present work the microstructure of an AlSi12CuMgNi (12 wt.% Si) alloy reinforced with two kinds of short fibers is mainly investigated by transmission electron microscopy (TEM). The reinforcements are polycrystalline $\delta$-$\text{Al}_2\text{O}_3$ fibers (Saffil), and fibers containing 50% $\text{Al}_2\text{O}_3$ and 50% $\text{SiO}_2$ (Fiberfrax). The diameter of both types of fibers varies from 1 to 3 $\mu$m. The materials were produced by *Mahle GmbH* in Stuttgart by infiltration of the molten alloy through a fiber preform.

The microstructure of the composite is characterized by a quasi-planar distribution of fibers immersed in an Al matrix with an inhomogeneous distribution of the solutes and of the precipitate phases. Al grains in contact with the fibers show a high dislocation density. The matrix contains large Si precipitates, which preferentially segregate at the fiber-matrix interface and often show a twinned structure. A needle-like Al-Ni phase and an Al-Cu-Mg-Si phase are also present in the matrix.

The Fiberfrax fibers partially transform to MgAl$_2$O$_4$ (spinel) during the production, owing to the reaction with Mg from the matrix. In the Saffil-reinforced composite, the spinel-forming reaction takes place only in the surface region of the fiber, where the preform binder ($\text{SiO}_2$) is present. After heat treatment at 350°C for 100 h (highly overaged condition), the matrix of the Saffil-reinforced composite shows needle-like coherent Al-Cu-Mg-Si precipitates.

Investigations of samples deformed in uniaxial compression experiments reveal cracks in the fibers and in the large Si precipitates close to the fibers, but not inside the Al grains or along the fiber-matrix interface, indicating a good adhesion. In the matrix, a subgrain structure with small-angle grain boundaries close to the fibers is observed. The size of the subgrains reduces to about 100 nm near the cracks in samples deformed to $\epsilon = 90\%$.

Small-angle scattering experiments are performed to investigate cavities or possible pores present at the interfaces between matrix and fibers. Calibrated small-angle scattering intensities are obtained in neutron and x-ray experiments. A comparison of the data indicates that the porosity of the fibers, with an amount of about 3 vol.% and an average pore radius of about 4 nm for both types of fiber, causes a predominant contribution to the scattering intensities. Any additional contribution from cavities and pores at the interface could not be detected.

Internal stresses and distortions of the matrix due to a different thermal contraction of matrix and reinforcement, are investigated with weak-beam imaging, convergent-beam electron diffraction (CBED), and large-angle CBED. Weak-beam images with different imaging conditions are compared with simulations of thickness fringes for varying thicknesses and lattice tilts (local orientations of the lattice). This method...
permits maps of sample thickness and lattice tilts to be obtained for the imaged grain. From the lattice tilt, shear strains or sample bending can be determined. The analysis of different regions of the sample reveals bending of the crystal especially close to the fibers and in thin places in the vicinity of the sample boundary. The sample bending is due to relaxation of internal stresses which are caused by the production process or during the preparation of the thin TEM sample.

A determination of the local lattice parameters by CBED is performed on Si precipitates. In the Al grains the dislocation density is too high for this kind of analysis. For the recognition of higher-order Laue zone (HOLZ) lines in CBED patterns, a method based on the Hough transformation is developed. The experimental data are compared with kinematic simulations corrected for dynamical line shifts. A least-squares fitting procedure considering the distances between the HOLZ line intersections, is developed to refine the lattice parameters. With a reduction of the number of unknown parameters or by combining the information from several different crystal orientations, it is possible to determine the strain values in the examined regions. The evaluated strains are of the expected order of magnitude, but the accuracy is affected by contamination and depends on the reproducibility of measurements at the same position, when more than one crystal orientation is used. The analysis performed with CBED is limited to places with optimal diffraction conditions, where the strain fields do not vary strongly. This complicates an evaluation of strain gradients close to the reinforcements. Theoretical considerations show that a unique determination of the entire set of six lattice parameters from the exact positions of six HOLZ lines is possible. However, the experimental accuracy needed for such a determination is, even under optimal conditions, limited by the width of the HOLZ lines, depending on the sample thickness.

In the large-angle CBED technique, an image of the sample is overlapped with a HOLZ pattern. Every position in the sample is correlated with the overlapping portion of the diffraction image. This technique permits the strain distribution in a region of the sample to be obtained from a single experimental pattern. The experimental images are taken with an energy filter in elastic mode, and the intensity variations are compared with kinematical and dynamical simulations. Thermal strain fields used for the simulations, are calculated analytically assuming a perfect cylindrical fiber in an infinite matrix. In particular, the distortion due to surface relaxation, corresponding to the actual situation in the samples prepared for electron microscopy, is considered. In the calculation of the large-angle CBED patterns, the local variations of the lattice parameters, with corresponding changes of the scattering vectors, are taken into account. The results obtained from comparing experimental and simulated patterns indicate that matrix strains in the sample are actually cau-
sed by thermal stresses. In addition to these long-range effects, local strains induced by small grains at the fiber boundary also contribute. These local distortions at the interface turn out to be important for the properties of the composite, since they can promote the formation of cracks.
Zusammenfassung

In der vorliegenden Arbeit wird die Mikrostruktur von zwei 20 Vol.% kurzfaserverstärkten Verbundwerkstoffen einer AlSi12CuMgNi (12 Gew.% Si) Legierung mittels Transmissionselektronenmikroskopie (TEM) untersucht. Die Fasern sind eine polykristalline δ-Al2O3-Faser (Saffil) bzw. eine polykristalline Mischfaser aus jeweils 50% Al2O3 und SiO2 (Fiberfrax). Der Durchmesser von beiden Fasern variiert zwischen 1 und 3 μm. Die verstärkten Legierungen wurde von der Firma Mahle GmbH in Stuttgart durch Infiltration der Metallschmelze in einen Faserformkörper hergestellt.


Untersuchungen von Proben, die in einachsigen Druckexperimenten verformt wurden, zeigen Risse in den Fasern und in den grossen Si-Ausscheidungen nah den Fasern, aber nicht innerhalb der Al-Körner oder entlang der Faser-Matrix-Grenzfläche, was auf eine gute Adhäision hinweist. In der Matrix entwickelt sich eine Subkornstruktur mit Kleinwinkelkorn grenzen in der Nähe der Fasern. Die Grösse der Subkörner ist in bis e = 90% verformten Proben in der Nähe dieser Risse auf ungefähr 100 nm reduziert.


In der Weitwinkel-CBED-Technik ist das Bild der Probe mit dem HOLZ-Beugungsbild überlagert. Jede Position in der Probe ist mit dem jeweils überlagerten Teil vom Beugungsbild korreliert. Mit dieser Technik können lokale Variationen von Verzerrun-
1 Introduction

A composite material is defined as a material consisting of two or more chemically distinct phases [1]. It may have properties that are not exhibited in the same form by the single constituents. In many cases, one component is a continuous phase in which a reinforcement (second phase) is distributed. Metal matrix composites (MMC) consist of a metal matrix with usually a ceramic reinforcement. The distinction of metal matrix composites from other two- or multi-phase alloys comes from the processing of the composites. In the production process the matrix and the reinforcement are mixed together. There are three basic types of reinforcement: continuous fibers, short fibers and particles. Particle or discontinuously reinforced composites are especially important as they are less expensive than continuous reinforced materials and they can be processed by conventional metallurgical techniques such as casting or powder metallurgy, followed by rolling, forging and extrusion [2].

Aluminum based composites reinforced by ceramic fibers offer improvements over the matrix alloy, combining the low mass density of aluminum alloys with an improved high stiffness and strength at high temperatures. The elastic modulus of the composite is higher than that of aluminum [3] and the resistance to thermal fatigue is improved by a lower crack growth rate [4]. These are important properties for automotive applications like for example local reinforcements of pistons or engine blocks. Alumina- and silica-based preforms used in a squeeze casting process seem to satisfy requirements of low production cost and reliability for the automotive industry.

The interfaces between the matrix and the fibers are very important for the behavior during the production process and the application of the composite. The main problems concerning the interfaces are the mechanical compatibility and adhesion between matrix and fibers, the elastic stresses in the vicinity of the fibers, the structural and microstructural changes under thermal and mechanical loads as well as the nature of induced plastic deformation processes. Previous studies of silica and alumina fiber reinforced Al alloys indicate that the formation and composition of reaction products at the fiber-matrix interface depend significantly on the composite processing conditions, the alloy composition, and the structure of the fiber surfaces [5, 6]. Chemical reactions at the interface can be important for the bonding between the matrix and the fiber. On the other hand, the reaction can change the microstructure of the fiber and thus its mechanical properties. In silica and alumina reinforced Al-Mg-Si alloys the formation of spinel (MgAl₂O₄) at the fiber surface was found by several authors [7, 8]. The reaction causes a microstructural change of the fibers and
a depletion of magnesium in the matrix associated with a reduced age hardenability of the composite [9, 10]. The metal matrix is likely to suffer several microstructural alterations during processing, and changes in its mechanical properties will result [11]. During cooling of the solidified composite the difference in the thermal expansion coefficients of matrix and fiber causes inhomogeneous internal stresses [12, 13]. In response to thermal or mechanical stresses, plastic deformation of the ductile metal matrix, cracking of the fibers, and failure of the fiber-matrix interface can take place [14]. The precipitation kinetics in the matrix is also influenced by the presence of reinforcements: internal stresses lead to a high density of dislocations near the fibers. The dislocations act as heterogeneous nucleation centers for precipitates in the matrix during aging [15]. As a consequence, the precipitate growth rate is accelerated and the diffusion in the matrix is enhanced, causing differences in the age hardening and thus in the mechanical properties of the composite [14].
2 The metal matrix composite

Many processes for fabricating metal matrix composites are available [16]. Most of them involve processing in the liquid state, while others comprise various deposition or powder techniques [17]. Casting involves liquid infiltration of a fiber bundle by the molten metal. In the "squeeze casting" or pressure infiltration, the melt is forced into the interstices of an assembly of short fibers, usually called a preform [18]. Isostatic pressure (about 100 MPa) is applied by a piston until solidification is complete. Squeeze casting obviates the requirements of good wettability of the reinforcement by the molten metal. Composites fabricated with this method show minimal reaction between the reinforcement and the melt because of short-time processing. However, the method presents two disadvantages: first, the applied high pressure leads to damage of the fiber preform, second, the matrix exhibits porosity caused by gas entrapping during processing [19]. Preforms are commonly fabricated by sedimentation of short fibers from a liquid suspension. Subsequently the liquid is drained off by compressing and heating the preform. To retain its integrity and shape, it is often necessary to use a binder. Various silica- and alumina-based mixtures have been employed as high temperature binders. The binding agent is normally introduced via the suspension liquid, so that it deposits or precipitates on the surface of the fibers, preferentially at fiber contact points, where it serves to lock the fiber array in a strong network [18].

The materials investigated in the present work are an AlSi12CuMgNi alloy (see Table 2.1) and two short-fiber reinforced metal matrix composites of this alloy with 20 vol.% of Saffil® or Fiberfrax®. Saffil is a nearly pure α-Al2O3 polycrystalline fiber with a silica content of about 3-4% and an average diameter of 2.8 μm. Fiberfrax is a fiber consisting of 50% α-Al2O3 and 50% SiO2 with an average diameter of 1.8 μm. The two reinforced alloys were manufactured by a squeeze casting process by Mahle GmbH, Stuttgart. SiO2 was used as binder to fix the fibers in the preform.

Table 2.1:
Composition of the AlSi12CuMgNi alloy (M124).

<table>
<thead>
<tr>
<th></th>
<th>Si</th>
<th>Cu</th>
<th>Mg</th>
<th>Ni</th>
<th>Fe</th>
<th>Mn</th>
<th>Zn</th>
<th>Ti</th>
<th>Al</th>
</tr>
</thead>
<tbody>
<tr>
<td>wt.%</td>
<td>11-13</td>
<td>0.8-1.3</td>
<td>0.8-1.3</td>
<td>1.3</td>
<td>0.7</td>
<td>0.3</td>
<td>0.3</td>
<td>0.2</td>
<td>balance</td>
</tr>
</tbody>
</table>

The three materials were provided by Prof. H.-J. Gudladt (Universität der Bundeswehr München, Germany). They were homogenized at 750 K for 10 h and quenched.
to room temperature in an oil bath. Some specimens of each material were subsequently heat treated for 100 h at 623 K to a highly overaged (HOA) condition. The pure alloy and the composites were mechanically polished for optical microscopy and scanning electron microscopy (SEM). Thin foils for transmission electron microscopy (TEM) examination were prepared by standard procedures using a Gatan Duo ion mill with liquid nitrogen cooling and a Gatan Precision Ion Polishing System (PIPS).

2.1 Optical microscopy and scanning electron microscopy

The three different materials are based on the same alloy, but the microstructure is strongly influenced by the presence of the fibers. Even with an optical microscope strong differences in the structures of unreinforced and reinforced alloys are clearly visible. In the alloy without reinforcement, rod- and needle-shaped precipitates, formed during the heat treatment and the cooling process, show a typical dendritic structure. The precipitates are distributed quite homogeneously in the matrix. Their length varies from 10 to 20 μm. An image analysis indicates an approximate volume fraction of 10% for both phases.

In the composite the fibers appear as black spots or ellipsoidal regions (see Fig. 2.1). The fiber distribution was inherited from the preform, and a nearly planar orientation of the fibers can clearly be seen by optical microscopy. The two types of precipitate observed in the pure alloy have the same volume fraction again, but they are considerably smaller and shorter in the composite. In Fig. 2.1 they appear gray


Figure 2.1:
and dark gray, and are preferentially distributed around the fibers, not following a dendritic scheme any more. Optical microscopy showed few differences between the Saffil and the Fiberfrax reinforced composites. In the latter, the precipitates are more finely and homogeneously distributed in the matrix except for some large precipitates which exceed a diameter of 100 μm. No differences between heat-treated and as-received samples were found by optical microscopy.

SEM examinations and qualitative energy dispersive spectroscopy (EDS) of the different precipitates were performed on a CamScan CS44. The dark gray phase observed by optical microscopy is Si. Large Si precipitates were found especially around the fibers and connect many of them forming a network. In SEM images, two additional phases are visible (see Fig. 2.2). A chemical analysis revealed an Al-Cu-Mg-Si phase

![Figure 2.2:](image)

Backscattered SEM micrograph of the unreinforced alloy with Si and intermetallic precipitates.

and an Al-Fe-Ni-Cu phase. Image analysis indicated approximate volume fractions of 6% and 4%, respectively. Small precipitates appearing black were also found. This phase with a volume fraction of 2-3% shows Al-Kα, Mg-Kα and Si-Kα peaks in the EDS spectrum but the intensity of the Al-Kα peak varies strongly with the position and the size of the particles, thus only Mg and Si can be definitely assigned to this phase. The phases in the unreinforced sample were also confirmed in the Saffil-reinforced composite. The Al content of the particles is lower than that of Mg and Si, but it could not be determined exactly, as the surrounding Al matrix also contributes to the EDS signal. The Al-Cu-Mg-Si and Mg-Si phases were not found
in the Fiberfrax-reinforced alloy. No differences between the images of as-received and heat-treated samples were found.

### 2.2 Transmission electron microscopy

The structure of the precipitates and the interfacial regions were investigated by TEM on a *JEOL* 200 CX and a *Philips* CM 30 equipped with a *Noran* Voyager X-ray microanalysis and digital imaging system, and a *Gatan* parallel detection electron energy loss spectrometer. As revealed by electron diffraction, the fibers are nanocrystalline and can easily be distinguished from the other phases in the matrix. In the thin TEM foils the fibers are cross-sectioned and appear as round or ellipsoidal with precipitates segregating at the interfaces to the matrix. Figure 2.3 shows a fiber embedded in the matrix with Si precipitates in the interfacial region.

![TEM micrograph of a fiber with twinned Si precipitates at the interface](image)

**Figure 2.3:**

TEM micrograph of a fiber with twinned Si precipitates at the interface

In the Al matrix a high amount of dislocations is present, with increasing density in the vicinity of fibers (see Fig. 2.4). The Al matrix tends to relax the thermal stresses, which build up during cooling after solidification, by plastic deformation. Conversely, in Si very few dislocations are present, but often twins are visible, especially in particles near the fiber-matrix interface. Figure 2.5 shows a high resolution
Figure 2.4:
Weak-beam image of dislocations close to the fiber-matrix interface in an Al grain of a Fiberfrax-reinforced sample.

electron microscopy (HREM) image along the [01\overline{1}] direction of a Si precipitate near a fiber. Twins on the (\overline{1}11) plane, starting at the interface, end in some case in the middle of the precipitate. A Burgers circuit around the side where the twin terminates, indicates that the resulting Burgers vector either vanishes or is parallel to the incident beam. The latter hypothesis is unlikely because in such a case the region around the core would be much more distorted by the Eshelby twist, and the image of the columns would be less clear. A vanishing sum Burgers vector can be explained considering the sequence of individual dislocations on consecutive (\overline{1}11) planes. The dislocations are partial dislocations with \( \frac{1}{6}[\overline{1}21] \), \( \frac{1}{6}[211] \) and \( \frac{1}{6}[\overline{1}12] \) Burgers vectors. An independent motion of the individual dislocations under the action of a stress is not possible. Thus this situation excludes thermal stress to be responsible for induced twinning.

Small particles in bright- or dark-field images of silicon were also observed. Because of the small size (10 nm and less) of these particles, the determination of the lattice constant from diffraction patterns and chemical analysis is difficult. Figures 2.6 and 2.7 show electron diffraction patterns along Si[110] and Si[100] with additional reflections coming from these small precipitates.

The small particles embedded in Si are well distinguishable in high-resolution images,
Figure 2.5:

HREM micrograph of Si twins close to a fiber.

Figure 2.6:

Diffraction pattern of Si[110] with additional spots from precipitates.
as they show typical Moiré patterns. Figure 2.8 shows a particle located in a twin interface. The existence of such coherent precipitates in Si has recently been reported by several authors [11, 8], but the phase has not yet been identified because of the difficult interpretation of the diffraction patterns.

The Al-Fe-Ni-Cu phase observed by SEM was further investigated by TEM, EDS and HREM. This needle-shaped phase grows into the Al matrix and shows no orientation relationship to the Al grains. The density of such precipitates varies, depending on the region, from isolated needles to compact aggregates as in Fig. 2.9. The diffraction patterns yield an orthorhombic structure with the lattice parameters $a = 23.3$ Å, $b = 7.44$ Å and $c = 13.79$ Å. Figure 2.10 shows a diffraction pattern along [031] with superstructure reflections indicating a particular periodicity. High resolution images taken along the same orientation show a complicated periodically modulated structure (Fig. 2.11).

For a quantitative analysis of the phase composition, EDS spectroscopy was used. Figure 2.12 shows an EDS spectrum of the Al-Ni-Fe-Cu precipitate. From the intensities of the different peaks, the ratio of the weight fractions between the components
Figure 2.8:
HREM micrograph along [110] of particle embedded in Si.

Figure 2.9:
Aggregation of needle-like Al-Fe-Ni-Cu precipitates.
Figure 2.10:
Diffraction pattern of the Al-Fe-Ni-Cu phase along [031].

Figure 2.11:
HREM micrograph along [031] of the Al-Fe-Ni-Cu phase.
can be calculated, neglecting absorption and fluorescence effects, from [20]

\[
\frac{C_A}{C_B} = k_{AB} \frac{I_A}{I_B} \tag{2.1}
\]

where \(C_A\) and \(C_B\) are the weight fractions of elements A and B in the sample, \(I_A\) and \(I_B\) are the intensities of the characteristic X-ray energy of these two elements, and \(k_{AB}\) is a constant of proportionality depending on the acceleration voltage. The latter can be measured from standards or calculated using [21]

\[
k_{AB} = \frac{Q_B \Omega_B a_B / A_B}{Q_A \Omega_A a_A / A_A} \tag{2.2}
\]

where \(Q_{A,B}\) is the ionization cross section, \(\Omega_{A,B}\) is the fluorescent yield, \(a_{A,B}\) is the ratio \(I(K_\alpha)/(I(K_\alpha) + I(K_\beta))\), and \(A_{A,B}\) is the atomic weight of the individual elements. For thick specimen the absorption is not negligible and the eq. 2.1 is modified to [21]

\[
\frac{C_A}{C_B} = k_{AB}^F \frac{I_A (\mu/\rho)^A}{I_B (\mu/\rho)^B} \frac{1 - \exp\left(-\frac{(\mu/\rho)^B}{\rho t \csc(\alpha)}\right)}{1 - \exp\left(-\frac{(\mu/\rho)^A}{\rho t \csc(\alpha)}\right)} \tag{2.3}
\]

where \(k_{AB}^F\) is the \(k_{AB}\) factor for an infinitely thin foil, \((\mu/\rho)^{A,B}\) is the mass absorption coefficient for X-rays of element A, respectively B in the specimen, \(\rho\) is the sample density, \(\alpha\) is the X-ray takeoff angle, and \(t\) is the specimen thickness.
EDS analysis involves a series of possible errors due to artifacts in the spectrum acquisition (e.g. interactions of electrons and X-rays with the sample away from the volume of interest, or with column components) or associated with the determination of $k_{AB}$ and the absorption or fluorescence corrections. The accuracy of $C_A/C_B$ is usually around 5%. The EDS analysis of the needle-like precipitates yields approximately Al-30 at.% Ni-8 at.% Fe for the main constituents. Si, Cu and Mn are present in small amounts. The lattice parameters are consistent with the $(Al,Si)_3(Ni, Cu, Fe, Mn)$ phase proposed by S. Wang et al. (1995) [8].

The other precipitates observed by SEM (Al-Cu-Mg-Si phase) were also investigated by EDS and the four elements were confirmed with varying concentrations. The analysis of the diffraction patterns yields an hexagonal structure with $a = 10.3$ Å and $c = 27.2$ Å. This phase may be related with the $Q(Al_2Cu_2Mg_8Si_7)$ phase (hexagonal structure $hP21$ with $a = 10.4$ Å and $c = 4.02$ Å) [22], which was often found in composites of similar Al alloys [23]. The difference in the lattice constant $c$ could be due to a different concentration of the elements in this phase and a different stacking sequence. Figures 2.13 and 2.14 show a diffraction pattern of the structure along the $c$ axis and its kinematical simulation, where the absolute intensity of the spots is arbitrary. The intensity ratio between spots in both figures is very similar, even though the differences in the intensities of the reflections in the experimental

![Diffraction pattern of Al-Cu-Mg-Si phase along [001].](image)

Figure 2.13:
pattern are reduced by multiple diffraction.

Figure 2.14:
Kinematical simulation of the diffraction pattern of the $Q(Al_4Cu_2Mg_8Si_7)$ phase along [001]. The weak spots around 000 are \{100\} reflections.

Investigations of the heat-treated samples have shown the presence of coherent needle-like precipitates in the matrix of the unreinforced and Saffil-reinforced alloys. The thickness of the precipitates varies from 50 to 150 nm, their length ranges from 500 nm to 2 mm. Diffraction patterns indicate a hexagonal structure with $a = 10.3$ Å and $c = 4.04$ Å. The lattice constants agree with the above-mentioned $Q(Al_4Cu_2Mg_8Si_7)$ phase, and EDS analysis exhibits the presence of Al, Si and Cu. Figure 2.15 shows the needle-like precipitates elongated parallel to the c-axis of the hexagonal structure and to the $<001>$ directions of Al. The orientation for the hexagonal basis are (see Fig. 2.16): $(410)_Q \parallel (100)_{AI}$ and $(350)_Q \parallel (010)_{AI}$. The same orientation relationships were found by other authors [24, 25, 26] in recent studies about the same kind of precipitate.

In the Fiberfrax-reinforced composite, small crystalline particles with a size between 500 nm and 1 μm were found near the surface of the fibers. They do not belong to the fiber, but they substitute parts of it. Diffraction patterns of this phase indicate a cubic symmetry with a lattice parameter of 8.08 Å. The phase was identified as spinel ($MgAl_2O_4$), in agreement with other investigations on composites of similar
Figure 2.15:
Coherent Al-Cu-Mg-Si precipitates parallel to Al[100].

Figure 2.16:
Diffraction pattern of Al[001] (main spots) with the overlapped pattern of the Q phase along [001].
composition [27, 6]. A Bragg reflection from one of these particles was selected for dark-field imaging (see Fig. 2.17). The image shows spinel with a white contrast appearing in the fiber near the interface. The small white spots inside and outside the fiber do not correspond to spinel but to other small crystalline phases accidentally selected (e.g. particles embedded in Si described above).

Figure 2.17:

Dark-field image of spinel particles in a Fiberfrax fiber in the Al matrix.

Possible reactions proposed by other authors for the formation of MgAl$_2$O$_4$ at the interface are

$$2\text{SiO}_2 + 2\text{Al} + \text{Mg} \rightarrow \text{MgAl}_2\text{O}_4 + 2\text{Si} \quad [10],$$

and

$$\begin{cases} 
\text{SiO}_2 + 2\text{Mg} \rightarrow 2\text{MgO} + \text{Si} \\
\text{Al}_2\text{O}_3 + \text{MgO} \rightarrow \text{MgAl}_2\text{O}_4 \quad [7] 
\end{cases}$$

In the Fiberfrax-reinforced composites, reactions with SiO$_2$, contained either in the binder or in the fibers, take place during the squeeze-casting process. As a consequence, the fibers and the interface lose their original structure. Additionally, the spinel formation reduces the magnesium content and the volume fraction of Mg-rich precipitates in the matrix [9]. This explains the absence of the Mg-Si and Al-Cu-Mg-Si phases in the Fiberfrax-reinforced samples.
From the nominal Mg content in the M124 alloy and from the approximate amount of spinel in the fiber found with TEM it is possible to evaluate the distribution of Mg in the composite. A rough calculation indicates that the Mg content in the matrix is high enough to form spinel occupying up to 25 vol.% of the fibers, which is slightly more than observed. Thus, a small amount of Mg might still remain dissolved in the matrix.

EDS linescans from the inner parts of the fibers into the matrix for the characteristic EDS peaks of Al-Kα, Si-Kα, O-Kα, and Mg-Kα have been performed (see Fig. 2.18). The original Fiberfrax fiber contains 50% Al2O3 and 50% SiC2, thus, at the beginning of the scan, in the center of the fiber, Al, Si and O occur with comparable intensity. Approaching the interface, the Mg content slowly increases with the presence of spinel. After reaching the maximal concentration in the spinel region, the intensity of the Mg peak decreases at the fiber boundary. In the matrix a high Al content with a constant low concentration of Mg, but still higher than inside the fiber, is observed. The Fiberfrax-matrix interface, as shown by the linescan, is not sharp and consists of a region of only spinel and intermediate zones with Fiberfrax and spinel, or spinel and matrix.

Figure 2.18:
EDS linescan through the Fiberfrax-matrix interface.

Electron Energy Loss Spectroscopy (EELS) served to further investigate the chemistry at the interface. For a quantitative analysis a ratio of the atomic fractions
$C_A$ and $C_B$ of two elements A and B can be determined, after background subtraction, from the integrated intensities of the ionization edges $I_A$ and $I_B$,

$$\frac{C_A}{C_B} = \frac{I_A \sigma_B}{I_B \sigma_A} \tag{2.5}$$

where $\sigma_{A,B}$ is the ionization cross section for the considered shell excitation. A quantitative EELS analysis is limited to sample thicknesses which are less than the inelastic scattering mean free path [20]. This limitation is caused by multiple inelastic scattering events, which contribute to the background, that an electron will undergo multiple inelastic scattering events, and thus contributes to the background rather than to a peak, increases. Quantitative uncertainties can derive from the calculated cross sections $\sigma_{A,B}$, that in general are accurate to only about ± 25% [20].

A part of the EELS spectrum, taken at the fiber boundary, and covering the Al-K and Mg-K edges is displayed in Fig. 2.19. Both edges are seen because these two elements are present in spinel. The fine structure of the Al edge exhibits a characteristic form due to the bonding with Mg and O in the MgAl$_2$O$_4$ phase.

![EELS spectrum of the Al-K and Mg-K edges in spinel.](image)

Figure 2.19:

EELS spectrum of the Al-K and Mg-K edges in spinel.

Figure 2.20 shows a part of a spectrum taken in the center of a Fiberfrax fiber, whose composition is a mixture of Al$_2$O$_3$ and SiO$_2$. The fine structure of the Al edge is different from that found in spinel. Thus, it is possible to distinguish whether the Al edge is produced in the reaction region or in the inner part of the fiber. A sequence
of EELS spectra acquired on a line from the interface to the center of the fiber indicates that Si exists only outside the spinel region and the fine structure does not change in the center of Fiberfrax fibers. The content of Si increases slightly from the boundary zone to reach a constant concentration in the middle of the fiber. Therefore, Si formed by the above-mentioned reactions most probably does not migrate towards the center of the fiber, but into the matrix.

![EELS spectrum of the Al-K and Si-K edges in Fiberfrax.](image)

Figure 2.20: EELS spectrum of the Al-K and Si-K edges in Fiberfrax.

In the Saffil-reinforced alloy, only the binder at the surface of the fiber contains SiO₂. Therefore, the spinel formation takes place in the surface region, where the binder covers the fibers as a homogeneous layer of about 100 nm thickness (see Fig. 2.21). HREM examinations of these layers reveal a complex microstructure with Al grains overlapping small spinel and Al₂O₃ particles with random orientation. Figure 2.22 shows a HREM image of a spinel particle oriented along [111] partially overlapped by other particles. The different orientations of overlapping grains produce a Moiré pattern. Owing to the reduced spinel formation in the case of Saffil fibers there is no Mg depletion in the matrix and no suppression of the formation of Mg-Si and Al-Cu-Mg-Si precipitates.

An EDS linescan across the Saffil interface shows a sharp Mg-rich range corresponding to the spinel layer around the fiber. Figure 2.23 displays a line scan across an interface between Saffil and a Si particle in the matrix. The second additional
Figure 2.21:
TEM micrograph of Saffil fiber cross section with reaction film (dark layer around the fiber) at the interface.

Figure 2.22:
HREM micrograph of spinel along the [111] direction. An overlapping grain on the left produces a Moiré pattern.
small peak of Mg is due to the formation of spinel from $\alpha$-Al$_2$O$_3$ within the largely predominant $\delta$-Al$_2$O$_3$ of the Saffil fiber. A model for this reaction has recently been proposed [28].

Figure 2.23:
EDS linescan through the Saffil-matrix interface. The intensity scale for Si and Al is vertically displaced to better recognize the single scans.

2.3 Transmission electron microscopy of deformed samples

The microstructure of one unreinforced and two Fiberfrax-reinforced samples was studied by TEM after macroscopic uniaxial compressive deformation to $\epsilon = 6\%$ (unreinforced), $\epsilon = 5\%$, and $\epsilon = 90\%$ (reinforced). A subgrain structure is observed both in the monolithic and in the reinforced alloy. In the latter, subgrains are concentrated around the fibers, where the dislocation density is high. After large deformation, the fibers are broken at several places, and some of the neighboring Si particles are fractured, too, as shown in Fig. 2.24. The crack does not enter the Al matrix but numerous dislocations are present to compensate for the large strains. Subgrains with a variable size from less than 100 nm up to 500 nm appear, which accomodate the strong deformation near broken fibers and Si particles (see Fig. 2.25). No decohesion at the interface of fiber and matrix occurs, confirming the good bonding between them. Subgrains are slightly tilted with respect to each other. Some grains in Fig. 2.25 are in a weak-beam condition, whereas others, with a slightly
Figure 2.24:
Dark-field image of a broken fiber with a crack in an adjacent Si particle.

Figure 2.25:
Weak-beam image with part of a broken fiber (below) and subgrain structure. Light grains are in the two-beam condition, whereas dark ones are out of Bragg condition. Grains with visible thickness contours are in weak-beam condition.
different orientation, are already in the two-beam case or not in Bragg condition at all. Dislocation networks are visible at the small-angle grain boundaries (see Fig. 2.26). The distance between these dislocations can be measured in projection on the TEM image. This allows the relative orientation of the subgrains to be estimated. From several dislocation networks, an average tilt angle of about 1° is found.

Figure 2.26:
TEM image of small-angle grain boundary with typical dislocation network.
3 Small-angle scattering

Small-angle scattering (SAS) with X-rays (SAXS) or neutron (SANS) are useful methods for the investigation of the microstructure of materials at a mesoscopic scale, typically in the range from about 1 nm to a few hundred nm. SAS is appropriate in materials science to investigate precipitation, decomposition, extended defects, and voids. The spatial dimension of these objects requires measurements at very small angles or scattering vectors.

3.1 Theory

The small-angle scattering intensity depends on the distribution of the scattering length in the sample in a range larger than the interatomic distance. The distribution can be expressed by a varying scattering length density $\rho(\vec{r})$. The intensity is

$$I(q) \propto \int_V \rho(\vec{r}) \exp(-i\vec{q} \cdot \vec{r}) d^3r$$

(3.1)

Depending on the characteristics of the sample and on the kind of information one needs about its microstructure (size of inhomogeneities, form, volume fraction, interfacial area, etc.) different models are assumed. A commonly used approximation in SAS is the two-phase model. In this case a system of two phases (matrix and particles) is assumed with two uniform scattering lengths densities $\rho_m$ and $\rho_p$. In the two-phase model, the integrated intensity $\bar{Q}$, considering an orientational average for $q$, is related with the volume fraction of the precipitates and the scattering length density difference $\Delta \rho = \rho_p - \rho_m$ [32]:

$$\bar{Q} = 4\pi \int_0^\infty I(q) q^2 dq = (2\pi)^3 (\Delta \rho)^2 C_p (1 - C_p)$$

(3.2)

$I(q)$ is the orientational average of the scattering intensity, and $C_p$ is the volume fraction of the precipitates. The parameter $\bar{Q}$ can be determined after extrapolation of the measured curve and numerical integration.

In the Porod law [31, 32], for a two-phase system with sharp interfaces the scattering intensity at large $q$ decreases asymptotically with $q^{-4}$ as

$$\lim_{q \to \infty} I(q) = K_P q^{-4} + B$$

(3.3)

with $K_P = 2\pi A (\Delta \rho)^2$. $K_P$ is the Porod constant, $A$ is the total interfacial area between the two phases and $B$ is the incoherent scattering background. If the contrast $(\Delta \rho)^2$ is given, then the area of the interfaces can be calculated.

The value of $\rho$ for neutrons is calculated as follows: $\rho = \nu b$, where $\nu$ is the atomic density and $b$ the scattering length. In the case of X-ray, $\rho = r_e \nu Z$, where $r_e$ is the electron radius and $Z$ the atomic number.
3.2 Background correction and intensity calibration

SAS intensities contain contributions from the sample and other objects, like for example pinhole, windows, sample container. Contributions not related to the beam are the electronic noise, cosmic rays and background from radiation sources and other instruments. For the corrections of these effects one needs three measurements: one with the sample, one without sample (but with the same measurement condition) and one for the "dark current". For the latter a strongly absorbing material replaces the sample (lead for x-rays and cadmium, gadolinium or boron for neutrons). The total intensity $I_T$ measured by the detector is given by

$$I_T = I_S + I_{DC} + \tau(I_{EB} - I_{DC})$$

(3.4)

where $I_S$ is the intensity scattered by the sample, $I_{EB}$ is the empty beam, $I_{DC}$ is the "dark current". The transmission coefficient $\tau$ of the sample is given by

$$\tau = \exp(-\mu d)$$

(3.5)

where $d$ is the thickness of the sample and $\mu$ its linear absorption coefficient.

The count rate $I_S$ for a detector pixel is further given by

$$I_S(x, y) = I_0 e(x, y) A \tau t \Delta \Omega(x, y) \frac{d\Sigma}{d\Omega}(x, y)$$

(3.6)

where $I_0 = $ flux of the primary beam $[s^{-1} \text{ cm}^{-2}]$, $e(x, y) =$ detector efficiency of pixel $(x, y)$, $A =$ cross section of the beam at the sample $[\text{cm}^2]$, $t =$ transmission coefficient of the sample, $\Delta \Omega(x, y) =$ solid-angle element of pixel $(x, y) [\text{sr}]$, $\frac{d\Sigma}{d\Omega}(x, y) =$ differential scattering cross section of the sample at pixel $(x, y) [\text{cm}^{-1} \text{sr}^{-1}]$.

The solid-angle element can be approximated, for small scattering angles, with $\Delta \Omega = a^2/d^2$, where $a$ is the pixel-size and $d$ is the sample-detector distance. If the efficiency of the detector is homogeneous over the whole surface, $I_0 e$ can be measured using a multiple-foil method or a secondary standard [30]. The transmission coefficient can be either directly determined through $\tau = I_S/I_{EB}$ (with $I_S$ measured for the unscattered beam), or can be calculated with eq. 3.5 given the thickness. The absolute differential scattering cross section $\frac{d\Sigma}{d\Omega}(x, y)$ can be calculated from eqs. 3.6 and 3.4.

For SANS calibration the incoherent scattering from vanadium or water is usually used. With vanadium the absolute cross section of the sample is

$$\frac{d\Sigma}{d\Omega}(x, y) = \frac{I_S(x, y) \tau_V t_V \Sigma^{inc}_V}{I_V(x, y) \tau_S t_S 4\pi}$$

(3.7)
where $\Sigma_{\text{inc}}^{\text{V}}=0.366 \text{ cm}^{-1}$ is the macroscopic incoherent scattering cross section of Vanadium, $\tau$ and $t$ are the transmission coefficient and the thickness, respectively. For water, multiple scattering has to be taken into account. A detailed discussion can be found in [29].

3.3 Experiment

Small-angle X-ray scattering experiments were conducted at the laboratory. The measurements were performed using a pinhole camera, a rotating anode X-ray generator and a 180 mm x 180 mm two-dimensional position-sensitive detector 2 m behind the sample. Monochromatic Mo-K$_\alpha$ radiation ($\lambda = 0.71 \, \text{Å}$) was employed and a range of scattering vectors within 0.2 and 4 nm$^{-1}$ was covered. The mechanically polished specimens (400 mm thick) were measured with the nearly planar oriented fibers perpendicular to the incoming beam.

Small-angle neutron scattering experiments were performed at the Risø National Laboratory, Roskilde, Denmark using a neutron wavelength of 10 Å and a two-dimensional position-sensitive detector. Sample-to-detector distances of 1 m and 6 m were employed to cover a large range of scattering vectors (0.03 up to 1.5 nm$^{-1}$). Figure 3.1 shows the geometry of the SANS instrument. Both orientations of the samples (5 - 5.5 mm thick), i.e. with the fibers oriented in planes parallel and normal to the neutron beam, were investigated. Furthermore, the fibers in the original state (before squeeze casting) were also measured. Boron carbide was used for “dark current “measurement. Water and vanadium were employed for calibration, and the results of both were in good agreement.

The two-dimensional SAXS patterns of the unreinforced alloy showed well-defined streaks with different orientations, depending on the position of the beam on the polycrystalline sample. Neutron measurements of the same specimen indicated a more isotropic scattering signal, as the beam diameter is larger. The streaks revealed the presence of coherent plate-like or needle-like precipitates in the matrix, with the smallest dimension of about 1 nm. The direction and length of the streaks depends on the illuminated region of the sample. Figure 3.2 shows a SAXS pattern of the unreinforced alloy with streaks.

A higher and more isotropic intensity was detected for reinforced samples. No important differences were observed between measurements with fibers oriented in planes parallel or perpendicular to the beam. Saffil fibers without matrix were also measured and the diagrams show almost the same absolute intensity as for the reinforced specimens. After background subtraction (see eq. 3.4) and absolute cali-
Figure 3.1:
Schematic of a SANS experimental setup.

Figure 3.2:
SAXS pattern of the unreinforced alloy.
bration (see eq. 3.6 and 3.7), the two-dimensional diagrams were radially averaged. A comparison between the averaged curve of the Saffil reinforced sample with that of the pure Saffil fibers shows a good overlap. The absolute intensity was calibrated for the same amount of fibers in both samples and the matrix intensity was subtracted. The pure fibers were weighted to determine the total scattering volume. For the composite sample the nominal concentration of 20 vol.% fibers were considered. Figure 3.3 shows the good agreement between the two curves.

The two comparable intensities indicate that the fibers are mostly responsible for the scattering signal in the Saffil- and Fiberfrax-reinforced composites. Assuming that the fibers contain pores, the scattering length density difference \( \Delta \rho \) to be considered for X-ray is \( \Delta \rho_{\text{Fiberfrax/Vacuum}} = 2.61 \times 10^{11} \text{ cm}^{-2} \) and \( \Delta \rho_{\text{Saffil/Vacuum}} = 3.35 \times 10^{11} \text{ cm}^{-2} \). For neutrons \( \Delta \rho_{\text{Fiberfrax/Vacuum}} = 4.5985 \times 10^{10} \text{ cm}^{-2} \) and \( \Delta \rho_{\text{Saffil/Vacuum}} = 5.736 \times 10^{10} \text{ cm}^{-2} \). The following scattering lengths were used: \( b_{\text{Al}} = 3.449 \times 10^{-13} \text{ cm} \), \( b_{\text{O}} = 5.803 \times 10^{-13} \text{ cm} \), \( b_{\text{Si}} = 4.149 \times 10^{-13} \text{ cm} \) [65]. The values of \( \nu \) for \( \text{Al}_2\text{O}_3 \) and \( \text{SiO}_2 \) contained in the fibers were calculated from \( \alpha-\text{Al}_2\text{O}_3 \) [59] and \( \text{SiO}_2 \) with hP12 structure [33].

Figure 3.4 displays the scattering cross section of the Saffil reinforced MMC measured with X-ray and neutrons, divided by the scattering contrast \((\Delta \rho)^2\) for pores in

![Figure 3.3:](image)

Calibrated SANS intensity of Saffil-reinforced composite (line) and pure Saffil fibers (dots).
Al₂O₃. Since the scattering contrast for X-ray and neutrons are different, the good overlap of the curves indicates that the assumption of pores in the fiber is justified. The strong increase at low q is due to (multiple) scattering from the fibers in the composite, whereas the slope at higher q originates from the pores.

Figure 3.4:
Scattering cross section of the Saffil-reinforced alloy divided by the scattering contrast of pores. Dots show the SANS data and open squares SAXS data.

The scattering curves of the Saffil- and Fiberfrax-reinforced alloy in the high q region fit well the Porod law (see eq.3.3) and information about the pores in both kinds of reinforcement can be obtained. The scattering intensity from the matrix has to be subtracted from the total intensity of the composite. The cross section is calibrated according to the 20 vol.% of reinforcement. As shown in Fig. 3.5 for the Saffil-reinforced composite, the Porod constant is found extrapolating the value of \( \frac{d\Sigma}{d\Omega} q^4 \) for \( q \rightarrow 0 \). The linear fit considers only the linear range significant for the pores.

The invariant \( \tilde{Q} \) is directly determined from the plot of \( \frac{d\Sigma}{d\Omega} q^2 \) over q with numerical integration. In the small q range, out of the scattering range due to the pores, the curve is approximated by a parabola following the \( q^2 \) slope. For large scattering
Figure 3.5:
Plot of $\frac{d\Sigma}{d\Omega} q^4$ vs. $q^4$ and extrapolation of $K_p$ at $q=0$.

Vectors, where no measured data are available, the integral $\int_0^\infty \frac{K_p}{q^2} dq = \frac{K_p}{q^2}$ is used. Figure 3.6 shows a plot of $\frac{d\Sigma}{d\Omega} q^2$ vs. $q$ for the Fiberfrax-reinforced MMC and the fitting parabola at low $q$.

Figure 3.6:
Plot of $\frac{d\Sigma}{d\Omega} q^2$ vs. $q$ for numerical determination of $\tilde{Q}$.

From eq. 3.2 the volume fraction of pores is determined. Combining eq. 3.2 with eq. 3.3 and neglecting the quadratic term $C_{\text{pores}}^2$ one obtains:

$$\frac{\tilde{Q}}{K_p} \approx (2\pi)^2 \frac{C_{\text{pores}}}{A}$$  \hspace{1cm} (3.8)

Assuming the pores to be spherical the ratio $\frac{C_{\text{pores}}}{A}$ becomes:

$$\frac{C_{\text{pores}}}{A} = \frac{4\pi}{3} \frac{R_{\text{pores}}^3}{A} = \frac{R_{\text{pores}}}{3}$$  \hspace{1cm} (3.9)
where \( R_{\text{pores}} \) is the radius of the spherical pore. Thus the size of the pores can be directly evaluated knowing \( \dot{Q} \) and \( K_P \):

\[
R_{\text{pores}} = \frac{3 \dot{Q}}{(2\pi)^2 K_P}
\]  

(3.10)

In the Saffil fibers an average pore radius of 4-5 nm with a volume fraction of about 3.5% was determined. In Fiberfrax fibers 1.5 vol.% pores of about 3 nm radius were found. The small-angle scattering investigations show that no considerable amounts of pores or cavities are present at the interface fiber-matrix, as is desirable for a good ceramic-metal adhesion. This is in good agreement with TEM investigations, where no pores at the interface were found.
4 Convergent-beam electron diffraction

4.1 Diffraction theory

The wave function describing the behaviour of an electron with energy $eU$ ($U$ is the acceleration voltage) moving in a crystal potential $\Phi(\vec{r}) = -eV(\vec{r})$ obeys Schrödinger equation

$$\Delta\Psi(\vec{r}) = -\frac{8\pi^2 me}{\hbar^2}(V(\vec{r}) + U)\Psi(\vec{r})$$

(4.1)

where $\hbar$ is the Planck constant, $e$ the positive elementary charge, and $m$ the relativistic mass of electron. The periodic potential $V(\vec{r}) = V(\vec{r} + \vec{t})$, where $\vec{t}$ is any translational vector of the crystal lattice, can be described as Fourier summation over all reciprocal lattice vectors $\vec{g}$ of the crystal

$$V(\vec{r}) = \sum_\vec{g} V_\vec{g} e^{2\pi i \vec{g} \cdot \vec{r}}$$

(4.2)

The wave function may be described as a sum of Bloch waves with amplitudes $\varphi_\vec{g}(\vec{r}) = \varphi_\vec{g}(\vec{r} + \vec{t})$ of all beams

$$\Psi(\vec{r}) = \sum_\vec{g} \varphi_\vec{g}(\vec{r}) e^{2\pi i (\vec{k} + \vec{g} + \vec{s}_g) \cdot \vec{r}}$$

(4.3)

Inserting eq. 4.3 in eq. 4.1 gives

$$\Delta\Psi = \sum_\vec{g} \left( \Delta\varphi_\vec{g} + 4\pi i (\vec{k} + \vec{g} + \vec{s}_g) \vec{\nabla}\varphi_\vec{g} - 4\pi^2 |\vec{k} + \vec{g} + \vec{s}_g|^2 \varphi_\vec{g} \right) e^{2\pi i (\vec{k} + \vec{g} + \vec{s}_g) \cdot \vec{r}}$$

(4.4)

$$= -\frac{8\pi^2 me}{\hbar^2} \left( \sum_\vec{g} V_\vec{g} e^{2\pi i \vec{g} \cdot \vec{r}} + U \right) \sum_\vec{g'} \varphi_{\vec{g}'} e^{2\pi i (\vec{k} + \vec{g} + \vec{s}_g) \cdot \vec{r}}$$

(4.5)

The Bloch functions vary slowly with $\vec{r}$ and the term $\Delta\psi_{\vec{g}}$ is negligible in comparison with $(\vec{k} + \vec{g} + \vec{s}_g) \cdot \vec{\nabla}\varphi_{\vec{g}}$. Furthermore for elastic scattering is $|\vec{k} + \vec{g} + \vec{s}_g|^2 = |\vec{k}|^2 = \frac{2meU}{\hbar^2}$.

It follows

$$\sum_\vec{g} (\vec{k} + \vec{g} + \vec{s}_g) \cdot \vec{\nabla}\varphi_{\vec{g}} e^{2\pi i (\vec{k} + \vec{g} + \vec{s}_g) \cdot \vec{r}} = \frac{2\pi ime}{\hbar^2} \left( \sum_\vec{g} V_\vec{g} e^{2\pi i \vec{g} \cdot \vec{r}} \right) \sum_\vec{g'} \varphi_{\vec{g}'} e^{2\pi i (\vec{k} + \vec{g} + \vec{s}_g) \cdot \vec{r}}$$

(4.6)
Since the components of the wave vectors $\vec{k} + \vec{g} + \vec{s}\bar{g}$ are essentially parallel to the beam direction, the left-hand side of eq. 4.6 can be approximated as $(\vec{k} + \vec{g} + \vec{s}\bar{g}) \cdot \nabla \varphi_{\bar{g}} \approx |\vec{k}| \frac{d\varphi_{\bar{g}}}{dz} = \frac{d\varphi_{\bar{g}}}{dz}$ and in the exponential terms $\vec{s} \cdot \vec{r} \approx s z$.

Eq. 4.6 has to be fulfilled for all $\vec{g} = \vec{g}' + \vec{g}''$ and comparing the coefficients of the exponential functions yields the many-beam Howie-Whelan equations [34, 35]

$$\frac{d\varphi_{\bar{g}}}{dz} = \sum_{\vec{g}'} i\pi \frac{2me}{\hbar^2} \lambda V_{\vec{g}'} e^{2\pi i(s_{\vec{g}' - \vec{g}'} - s_{\vec{g}})z} \varphi_{\vec{g}' - \vec{g}}$$

(4.7)

The terms with the Fourier coefficients of the periodic potential can be abbreviated defining the extinction distances $\xi_{\bar{g}} = \frac{\hbar^2}{2me\lambda V_{\bar{g}}} \approx \frac{\hbar^2}{2me\lambda V_{\bar{g}}}$, where $\lambda_c$ is the wavelength of the electron in the crystal.

The absorption can also be incorporated in eq.4.1 and eqs.4.7 introducing an additive imaginary lattice potential $V'_{\bar{g}}$: $V_{\bar{g}} \rightarrow V_{\bar{g}} + iV'_{\bar{g}}$. Consequently the extinction distance $\frac{1}{\xi_{\bar{g}}}$ is replaced by $\frac{1}{\xi_{\bar{g}}'} + \frac{1}{\xi_{\bar{g}}''}$, where $\xi_{\bar{g}}'$ is the mean absorption distance and $\xi_{\bar{g}}''$ are anomalous absorption distances.

Eq. 4.7 can be written in matrix form [36]

$$\frac{d\varphi}{dz} = \mathbf{A}\varphi \quad \text{with} \quad A_{jk} = \frac{i\pi}{\xi_{j-k}} e^{2\pi i(s_{j-k})z}$$

(4.8)

The components of the vector $\varphi$ are the functions $\varphi_{\vec{g}}$. $\xi_{j-k}$ are the extinction distances for scattering from beam $k$ to beam $j$ and $s_j$ the deviation parameter for the $j$-th function. With the substitution $\varphi_j = e^{-2\pi is_j z} \psi_j$, we obtain

$$\frac{d\psi}{dz} = \mathbf{M}\psi \quad \text{with} \quad M_{jk} = \frac{i\pi}{\xi_{j-k}} + \delta_{jk}2\pi is_j$$

(4.9)

The solution of eq. 4.9 can be written as

$$\psi(z) = \lim_{N \to \infty} \left( \mathbf{E} + \frac{\mathbf{M}z}{N} \right)^N \psi(0) = \sum_{n=0}^{\infty} \frac{(Mz)^n}{n!} \psi(0)$$

(4.10)

where $\mathbf{E}$ is the unit matrix. By solving the eigenvalue equation $\mathbf{M} = \mathbf{TDT}^{-1}$ with $\mathbf{T}$ containing the eigenvectors of $\mathbf{M}$ and $\mathbf{D}$ containing the eigenvalues of $\mathbf{M}$ as diagonal elements one obtains

$$\psi(z) = \sum_{n=0}^{\infty} \left( \frac{D^n z^n}{n!} \right) \psi(0) = \mathbf{T} \sum_{n=0}^{\infty} \left( \frac{D^n z^n}{n!} \right) \mathbf{T}^{-1} \psi(0) = \mathbf{T} \mathbf{L} \mathbf{T}^{-1} \psi(0)$$

(4.11)
In this equation the elements of $T$ represent the amplitudes of the Bloch waves. The matrix $L$ consists only of diagonal elements of the form $e^{i\kappa z}$. The imaginary part of the eigenvalues $\kappa_l$ is the wave vector of the corresponding Bloch wave.

In a deformed crystal the same derivation starting from the Schrödinger equation is possible. If the crystal lattice is distorted, the potential at $\vec{r}$ will change from $V(\vec{r})$ to $V(\vec{r} - \vec{u}(\vec{r}))$. The potential in the distorted crystal can thus be written as a Fourier series

$$V(\vec{r}) = \sum_{\vec{g}} V_{\vec{g}} e^{-2\pi i \vec{g} \cdot \vec{u}(\vec{r})} e^{2\pi i \vec{g} \cdot \vec{r}}$$  \hspace{1cm} (4.12)

The Howie-Whelan equations 4.9 can be formulated with a modified matrix [36]

$$N_{jk} = \left( \frac{i\pi}{\xi_{j-k}} + \delta_{jk} 2\pi is_j \right) \exp(2\pi i (\vec{g}_j - \vec{g}_k) \cdot \vec{u})$$ \hspace{1cm} (4.13)

Since $\vec{u}(\vec{r})$ changes along the integration direction $z$, eq. 4.10 can be applied only on small slices, where the matrix $N$ is assumed to be constant. The latter may be rewritten as [38]

$$N = R^{-1} M R$$ \hspace{1cm} (4.14)

We apply the same procedure as in eq. 4.11 for a slice of thickness $\Delta z$. Introducing again the matrix of eigenvectors $T$ and the diagonal matrix $D$ of eigenvalues $\kappa_l$ defined by $M T = D T$ yields [39]

$$\bar{\psi}(\Delta z) = \sum_{n=0}^{\infty} \left( \frac{(N \Delta z)^n}{n!} \right) \bar{\psi}(0) = R^{-1} T \sum_{n=0}^{\infty} \left( \frac{(D \Delta z)^n}{n!} \right) T^{-1} R \bar{\psi}(0)$$ \hspace{1cm} (4.15)

To determine the wave function at the thickness $z$, eq. 4.15 has to be applied for $N$ slices $\Delta z = \frac{z}{N}$ with $N$ different matrices $R$. With the diagonal matrix $L$ containing the elements $\exp(\kappa_l \Delta z) = \exp(\kappa_l \frac{z}{N})$ one obtains [38]

$$\psi(z) = \prod_{n=0}^{N-1} \left( R^{-1} \left( \frac{z}{N} \right) T L T^{-1} R \left( \frac{z}{N} \right) \right) \psi(0)$$ \hspace{1cm} (4.16)

The eigenvalues and eigenvectors are constant for all steps. Only the diagonal matrices $R$ and $R^{-1}$ change in the iteration, whereas the matrices $T$ and $T^{-1}$ are constant, and the diagonal matrix $L$ depends only on the actual step size.
4.2 Convergent-beam electron diffraction (CBED)

In the convergent-beam technique, originally proposed by Kossel and Möllenstedt [37], the electron beam is focused on the sample to a very small probe with a few mrad convergence. If the electron beam convergence increases, the diameters of the circular spot of the primary beam and of the Bragg diffraction spots increase. For a point-like electron source each point in a circle corresponds to one distinct direction of incidence in the illumination cone. The intensities within the primary beam disc and the Bragg diffraction discs vary owing to the variation of the excitation errors. The intensity distribution inside any Bragg diffraction disc is equivalent to the rocking curve calculated by the dynamical theory. The high beam convergence allows the excitation of reflections in higher-order Laue zones (HOLZ), and three-dimensional information about the crystal symmetry can be obtained. So called HOLZ lines arise when electrons within the incident beam at the correct Bragg angle for diffraction by a HOLZ plane are scattered to high angles, creating a bright line through the HOLZ spot and leaving a dark line in the central spot (see Fig. 4.1). These HOLZ lines stem from planes with much larger Bragg angles than in conventional diffraction patterns, so they are very sensitive to changes in lattice parameter. Therefore the CBED technique may be used for lattice parameter [42, 43] or crystal symmetry determination [40, 41].

Figure 4.1:
Scheme showing the principle of the CBED technique.
4.3 Large-Angle (LA)-CBED

The LACBED method has been originally developed by Tanaka [44] to obtain an angular view larger than the Bragg angle without overlapping between primary spot and diffracted spots. The electron probe is focussed above or below the sample and the objective lens is adjusted so that an image of the electron source, in the plane of the selected area aperture, for the incident and the diffracted beams is formed. The aperture can then be used to isolate one source image and so prevent other diffracted beams from contributing to the final pattern. The electron optical situation is illustrated by the ray diagram in Fig. 4.2. The electron probe is overfocused by $\Delta f$ and the diffracted beam (broken lines) can be intercepted by the selected area (SA) aperture. Because the source images are small at the crossover, the illumination cone can be opened up to a semiangle which is larger than the Bragg angle. A large area of the sample is illuminated by the unfocused beam and different regions of the specimen contribute to different parts of the diffraction pattern. Instead of adjusting
the objective lens current it is also possible to vary the eucentric height. The latter parameters and the probe focus may be chosen to have an appropriate angular range compatible with the desired illuminated portion of the sample. Of course, the condition of non-overlapping spots with the use of the selected aperture limits the possible combinations between convergence and area. A small selected area aperture together with a large defocus minimize the contribution of inelastic scattering [45].

In LACBED patterns, information from both real and reciprocal space is present. The HOLZ lines are superposed to a shadow image of the illuminated area in the sample. It is not possible to have both a sharp image and sharp HOLZ lines. A sharp real image is accompanied by blurred lines or vice versa depending on the imaging conditions. The latter situation corresponds to LACBED, where the microscope is usually adjusted in diffraction mode, i.e. the screen is conjugate to the focal plane of the objective lens or diffraction plane. The angular width of the lines is limited only by the diffraction limit imposed by the objective lens since the diffraction pattern is in focus, while the spatial resolution is limited by the cross-over size of the electron probe, as shown in Fig. 4.2. Thus there are contributions to a particular line from an area at least as large as the defocused probe and extended throughout the thickness of the sample. Experimentally, however, LACBED patterns are usually obtained at some intermediate focus condition, giving compromise performance. The position and the intensity of a line in a certain point in the pattern is directly correlated with the superposed point of the sample image. Thus it is possible to investigate defects or local strain distributions. Many authors have studied the effects of dislocations or stacking faults in LACBED patterns [46, 47, 48]. Some authors also investigated strain fields around particles [50] or in the vicinity of interfaces [49].

A method similar to LACBED called Convergent Beam Imaging (CBIM) has been developed by Humphreys et al. [51]. In this technique the electron beam is focused onto the specimen plane, i.e. the screen is conjugate to the image plane (see Fig. 4.2). Thus the angular width of HOLZ lines depends on the cross-over size of the probe, while (since the sample is in focus) the spatial resolution is given by the normal performance of the microscope. This corresponds to the case with a sharp image and blurred lines. Figure 4.2 shows that a HOLZ line (continuous line on the image plane) is blurred owing to the finite size of the cross-over. To select only the incident beam the objective aperture has to be inserted as for bright-field images. The selected area aperture is not used in this configuration and the diffracted beams may overlap.
5 Analysis of sample bending

5.1 Weak-beam contrast

From the Howie-Whelan equations (see eq. 4.7) for the two beam case one obtains the following intensity for the diffracted beam:

\[ I_{\bar{g}} = \left( \frac{\pi z}{\xi_{\bar{g}}} \right)^2 \sin^2 \left( \frac{\pi z}{s_{\text{eff}}} \right) \]

where \( s_{\text{eff}} \) is the effective excitation error

\[ s_{\text{eff}} = \sqrt{s^2 + \frac{1}{\xi_{\bar{g}}^2}} \]

The intensity \( I_{\bar{g}} \), the dark-field (DF) image intensity, behaves in a complementary manner to the direct beam used for bright-field (BF) images and is periodic in the two independent variables \( z \) and \( s_{\text{eff}} \). If \( s \) (and hence \( s_{\text{eff}} \)) remains constant while \( z \) varies (as in a wedge-shaped sample), then one observes an oscillation of the BF- or DF-intensity as a function of thickness, i.e. so-called thickness fringes. On the contrary, if \( z \) remains constant while \( s \) changes, the image shows so-called bending contours. In fact, a change in the deviation parameter \( s \) is equivalent to a local change of the crystallographic orientation with respect to the incident beam direction, corresponding to a local bending. At Bragg condition the thickness difference between two fringes has the value of the extinction distance \( \xi_{\bar{g}} \). In the general case with more than two beams, the many-beams dynamical formalism shown in chapter 4 has to be used. A local change of the deviation parameter \( s \) is then described by the expression \( s + \bar{g} \cdot \frac{dz}{dx} \) where \( \bar{u} \) is the local displacement. Using this formalism it is possible to calculate thickness fringes for a wedge-shaped sample with different lattice tilt \( \frac{dz}{dx} \) for different imaging conditions. The latter depend on the deviation parameter of the reflection selected in the DF image, and thus on the orientation of the crystal relative to the incident beam. In the weak-beam (WB) technique [57] a large \( s \) is chosen, so that \( 1/\xi_{\bar{g}} \) becomes negligible in \( s_{\text{eff}} \). The effective extinction distance \( \xi_{\text{eff}} = 1/s_{\text{eff}} \) is then much smaller than for dark-field imaging with \( s \approx 0 \), and the thickness fringes are very sensitive to a thickness change or foil bending. For WB images the notation \( (\bar{g}, n\bar{g}) \) is used, where \( n\bar{g} \) is the position in reciprocal space where the Ewald sphere cuts the line in reciprocal space determined by the direction of the \( \bar{g} \) vector. So \( n \) is not an integer if no reflection is exactly excited. To find out the values of \( n \), one observes the position of \( \bar{g} \)-Kikuchi lines relative to the fixed diffraction spots. The \( \bar{g} \)-Kikuchi band is always on the median point between the \( \bar{0} \) and the \( n\bar{g} \) reflection. Thus, if the \( n\bar{g} \) reflection is excited, then the \( \bar{g} \)-Kikuchi band
will be centered on the \( \frac{n}{2} \bar{g} \) spot, i.e. the \( \bar{g} \)-Kikuchi line is in the position \((n/2 + 1/2)\bar{g}\) while the \(-\bar{g}\) line of the Kikuchi band is in the position \((n/2 - 1/2)\bar{g}\).

A method proposed by Heinrich et al. [39, 56] uses WB images taken with different imaging conditions to determine shear strains or bending. Every point on the WB image may be related to a certain thickness and a lattice tilt just evaluating the thickness fringes in two different WB conditions. Since the product \( s + \bar{g} \cdot \frac{d\xi}{dz} \) is important for the image contrast, the displacement component parallel to \( \bar{g} \), i.e. a tilt \( \frac{d\xi}{dz} \), can be determined. The number of fringes separating a point from a reference position (usually the hole in the sample) may be used to determine the thickness at every point on the sample, if \( \xi_{\text{eff}} \) is known and no change of lattice tilt exists. If bending is present, it is necessary to take an image under another condition and to count again the fringes for the same position. Knowing the number of fringes for the two conditions, there is only one combination of thickness and lattice tilt for that position on the sample. The solution can be found using calibration maps from many-beam dynamical simulations with varying thickness and \( \frac{d\xi}{dz} \). For all positions between the fringes the values are interpolated. In WB the fringes are very close to each other so that a quite precise map of thickness and tilt over the sample can be obtained. The expression \( \frac{d\xi}{dz} \), for a zero-order Laue zone (ZOLZ) reflection \( \bar{g} \), corresponds to the term \( \frac{d\xi}{dz} \) in the expression relating the shear strain and displacement \( \epsilon_{xz} = \frac{1}{2}(\frac{dn}{dz} + \frac{dx}{dz}) \). The case with \( \frac{dn}{dz} = -\frac{dx}{dz} \) indicates a pure bending of the sample, whereas \( \frac{dx}{dz} \) gives a pure shear strain, if \( \frac{dn}{dz} = 0 \). If bending of the sample can be neglected, the analysis of the fringes provides a good method for determining shear strain fields.

### 5.2 Determination of sample thickness with CBED

To determine the thickness of the sample in a reference point, in a region far from a hole, the CBED method [58] can be used. As indicated by eq. 5.1 the intensity modulates for a fixed thickness as a function of the effective deviation parameter \( s_{\text{eff}} \). In CBED patterns, intensities for the different parameters \( s \) can be simultaneously recorded. The modulation is visible in so-called Kossel-Möllenstedt (KM) fringes, accompanying ZOLZ lines in the form of side maxima with decreasing intensity. The condition for these side maxima in the diffracted disk is \( z s_{\text{eff}} = n \), with \( n \) integer. Using eq. 5.2 one obtains

\[
\frac{s^2}{n^2} + \frac{1}{n s^2 g^2} = \frac{1}{z^2}
\]  

(5.3)
A plot of \((s/n)^2\) against \(1/n^2\) gives \(1/z^2\) as the intercept, and hence the thickness. The slope gives \(1/\xi^2\) from which \(V_g\) can be obtained. The value of \(s\) for each KM fringe can be found using

\[ s \approx \frac{\Delta x}{x} \frac{g^2 \lambda}{x} \tag{5.4} \]

where \(x\) is the distance between the centers of the incident disk and the diffraction disk. The length \(\Delta x\) is the separation distance of the KM fringes.

### 5.3 Bending in the fiber-reinforced composite

A fiber-reinforced sample has been investigated using the WB method. Regions with small thickness or close to fibers (or large precipitates) show particular fringe patterns under WB condition. Modifying the imaging conditions the patterns do not change in a regular way as expected from simple thickness fringes. Owing to ion polishing, the thickness in the sample usually varies from the hole to the thickest places with a regular increase. Variations of the fringe pattern on a short scale in the sample indicate an additional local change of the crystal orientation, i.e. a lattice rotation. Such a tilt may be determined comparing two images of the same region at the opposite WB conditions, using \((\vec{g}, n\vec{g})\) and \((-\vec{g}, -n\vec{g})\). If the fringes do not match, an additional rotation is present. Figure 5.1 shows an Al grain under \((\vec{g}, 13\vec{g})\) condition with \(\vec{g} = (111)\). The chosen region is the same as in Fig. 2.21. In the lower left part without fringes, a precipitate lies at the fiber interface. The

![Figure 5.1:](image)

Al grain imaged with \((\vec{g}, 13\vec{g})\) condition.
region in the lower right corner indicates a particular fringe pattern. To analyze the lattice tilt in the grain, another image with $(-\hat{g}, -15\hat{g})$ was taken. Both images were transformed into diagrams with the positions of the fringes. The value between them were interpolated and every point in the image was associated to a number of fringes. The experimental fringe patterns were compared with simulated ones to determine the lattice rotation. Figure 5.2 shows calculated thickness fringes in a wedge shaped sample, with thickness along the horizontal axis changing continuously from 0 to 200 nm. On the vertical axis the lattice tilt $\frac{du_z}{dz}$ varies continuously from -0.02 to 0.02. The WB condition in the center of the image is $(\hat{g}, 13\hat{g})$ with reflection $\hat{g} = (111)$ of Al. Combining the data from both images taken at different WB conditions, one obtains a map of thicknesses and lattice tilts of the considered region as shown in Figs. 5.3 and 5.4. The hole and the precipitate are black in both figures. The thickness varies regularly from the hole up to the thickest place on the left margin with about 160 nm. The lattice rotation changes strongly close to the precipitate and to the hole. The line with zero tilt is arbitrarily chosen, because only the relative rotation is important. In this simulation the region with zero rotation is the place where the two imaging conditions were determined. The precipitate at the fiber interface is responsible for the lattice distortion. However, its influence to the bending of the

Figure 5.2:
Calculated thickness fringes for $(\hat{g}_{(111)}, 13\hat{g}_{(111)})$ condition in Al. Thickness from 0 to 200 nm and $\frac{du_{(111)}}{dz}$ from -0.02 to 0.02.
Figure 5.3:
Thickness map of the region in Fig. 5.1. The line distances indicate height variations of 8 nm.

Figure 5.4:
Map of lattice tilt $\frac{du_{111}}{ds}$ (in $10^{-3}$) for the region in Fig. 5.1.
sample is strong enough only in the thinnest region close to the hole. The hypothesis of bending is more plausible than that of pure shear strain, since no shear source is present. In the thick region, stress induced from the particle is also present, there it cannot be relaxed by bending. The most stable situation for the sample with that thickness and internal stress is a uniform strain across the thickness. In the bending case strains are opposite on the top and on the bottom of the sample. The strong variation directly around the hole is questionable, since at small thicknesses the errors resulting from the positioning of the fringes increase. At higher thickness the weight of every single fringe is reduced, and thus the relative error as well. In Fig. 5.5 a very large Si grain surrounded by several Fiberfrax fibers is shown. The imaging condition is \( (\vec{g}, 5.5\vec{g}) \) with \( \vec{g} = (111) \) in a \([123]\) zone axis. In this case the hole is not visible in the image, and for the calibration of the thickness, CBED patterns are used. Taking another image with \((-\vec{g}, 7\vec{g})\) condition and combining them with the same procedure as explained above, one obtains a thickness and a lattice tilt map of the region as shown in Figs. 5.6 and 5.7. The region surrounding the grain is displayed in black. Approximately in the center of the grain the thickness is 170 nm and increases close to the boundary, every line indicating a 30 nm variation. The tilt map shows an almost uniform bending along the whole grain. The bending in this case is unlikely to be caused by relaxation of the foil, as it is too thick. The surrounding fibers have an effect on the lattice distortion that was already present.

Figure 5.5:
Si grain imaged with \((\vec{g}, 5.5\vec{g})\) condition.
Figure 5.6:
Thickness map of the region in Fig. 5.5. The line distances indicate height variations of 30 nm.

Figure 5.7:
Map of lattice tilt $\frac{dn_{11\bar{1}}}{dz}$ (in $10^{-3}$) for the region in Fig. 5.5.
before thinning of the sample. The internal stresses are accommodated through such a bending.

The tilt values found by WB analysis can be confirmed by using CBED or LACBED. Figures 5.8 and 5.9 show CBED patterns approximately along the [113] zone axis at 230 kV taken in the positions A and B, on the Si grain of Fig. 5.5. The patterns were taken at liquid nitrogen temperature to reduce the contamination of the sample.

![Figure 5.8:](image)

[113]-CBED pattern of Si grain at the position A of Fig. 5.5.

The HOLZ line pattern in the central disc changes according to the orientation of the crystal. If the grain is bent, the orientation relative to the incident beam slightly varies at different positions. One can assume in a first approximation that the line pattern does not change, i.e. the relative positions of the lines, one compared to the other, remain constant. To determine the lattice rotation, one measures the position change of one intersection between two lines in relation to a fixed point, for instance, the center of the disc. To convert the measured distance into an angle, a comparison with a known diffraction angle (for instance between two spots or two line intersections) is required. In the case mentioned above, one measures a tilt for the (110)-plane of about 4 mrad between position A and B. The [110] direction points perpendicular to the ZOLZ line (broad black line). In an equivalent way the rotation can be considered with a tilt of the zone axis in the plane containing [110] and [113]. The values obtained with WB simulations of the same region were related...
Figure 5.9:

[113]-CBED pattern of the Si grain at the position B of Fig. 5.5.

to the (11\overline{1}) plane in the [123] zone axis. For a rough estimate the same zone axis for the WB and the CBED case may be assumed. Using the rotated [113] zone axis, one finds that the tilt of the (11\overline{1}) plane is about 3 mrad, which is comparable to the results from the WB images.

The same Si grain was also investigated with LACBED. Figure 5.10 shows a pattern with very large convergence angle, taken without the selected area aperture. The central and the diffracted spots overlap, thus both defect and excess lines of the ZOLZ are visible. Owing to the large convergence, several poles around [7 10 9], connected by ZOLZ lines, can be seen similar as to a Kikuchi map. The displacement between excess and defect lines is due to bending of the sample. As shown in Fig. 5.11, the Bragg condition is satisfied for the right and the left beam, but since the sample is bent, the diffracted beam does not follow the same trajectory as the undiffracted beam on the opposite side, and in the diffraction plane the two lines do not coincide. Depending on the position of the primary-beam cross-over, i.e. above or below the sample, the defect lines are inside the excess ones or vice-versa. The same change is also obtained for an inverted bending of the sample. The displacement of one excess-defect couple with respect to the other may be measured and compared with a known value, as for instance the distance between a defect and its excess line, which is proportional to the Bragg angle. Examining the displacements in Fig. 5.10
Figure 5.10:
LACBED pattern with overlapping spots, taken around the [7 10 9] pole.

Figure 5.11:
Ray diagram for LACBED of a bent sample.
one obtains lattice tilts of about 2 mrad, consistent with the above considerations. Figure 5.12 shows the pole [7 10 9] overlapped in a LACBED pattern with a region very close to the central fiber in Fig. 5.5.

![Figure 5.12: LACBED pattern with [7 10 9] pole in a region very close to the fiber.](image)

The displacement for the ZOLZ lines (123) and (123) has changed compared with that in Fig. 5.10, i.e. in this region the sample is differently bent, with a larger tilt $\frac{du_{(123)}}{dz}$. This tilt change is compatible with the large increase of $\frac{du_{(123)}}{dz}$ found in the same region in the map of Fig. 5.7, considering that the projections of the distortions on planes perpendicular to the directions [123] and [111] do not differ too much. Furthermore the vicinity of the fiber causes a splitting of the line, owing to internal stresses not directly related to bending. This case will be discussed in chapter 8. Figure 5.13 shows the [7 10 9]-pole on the opposite side of the grain close to another fiber on the left. The displacement of defect and excess lines is like in Fig. 5.10, confirming again the result obtained with the WB analysis.

Using LACBED with a minor beam convergence and illuminating a smaller area of the sample, more accurate patterns can be obtained, with almost straight defect lines, without the disturbing presence of the excess intensity. The pattern in Fig. 6.5 was taken approximately along the [547] axis of a Si grain at the interface of a fiber, with an acceleration voltage of 120 kV. To evaluate the lattice distortion, another image with the same condition but in a position closer to the fiber has also been considered. Both images were digitized using the Hough transformation (see next chapter). The superimposed patterns are shown in Fig. 5.14. The dashed line corresponds to the pattern closer to the fiber. The latter is situated on the
Figure 5.13:
LACBED pattern with \([7 10 9]\) pole in the region on the opposite side of the grain, with fiber on the left (see Fig. 5.12).

Figure 5.14:
Superposition of two LACBED patterns along approximately the \([547]\)-pole. Dashed and solid lines show the pattern for a position near and far from the fiber.
left side of the pattern. The closer to the fiber, the more displaced are the lines of
the two patterns with respect to each other. Furthermore the "splitting" is small
for lines with directions almost perpendicular to the lateral margin, i.e. pointing
to the fiber center. In contrast, the vertical lines are strongly split. This indicates
that the fiber actually influences the surrounding matrix and the latter is bent in
radial direction in its vicinity. Additional distortions are visible near the upper-left
corner of the superposed patterns. A detail of Fig. 5.14 is redrawn for both patterns
in Fig. 5.15. The line intersections are clearly different. A simple bending is not
enough to displace the intersection points so strongly. This situation demands an
accurate analysis of the position of every line, as will be shown in the next chapter.
A determination of the lattice parameters of the crystal at the considered place is
needed to determine its strain condition.

![Diagram](image)

**Figure 5.15:**
Detail on the upper left corner in Fig. 5.14, showing different line
intersections.
6 Determination of lattice parameters

As explained in chapter 4, CBED provides a good method for the determination of local lattice parameters. The strain is related to the Bragg angle according to \( \Delta a/a = \Delta \theta/\theta \). The accuracy of the strain determination increases using HOLZ reflections with high diffraction angles instead of ZOLZ reflections. Furthermore, HOLZ reflections contain a component relative to the third dimension (along the zone axis), which is useful to study the spatial symmetries.

6.1 Geometry of HOLZ lines

In the kinematic approximation, the position of HOLZ lines can be found from geometrical considerations. A HOLZ line position is the locus of the Bragg condition for a HOLZ reflection \( \vec{h} \). The lines occur in pairs, with an increased intensity (in comparison with the intensity for similar wave vectors for which the Bragg condition is not exactly fulfilled) in the outer HOLZ ring (excess line) and a corresponding lowered intensity in the incident-beam disk (defects line). Figure 6.1 shows a simplified scheme of HOLZ line geometry. The two-dimensional representation contains the zone axis and is perpendicular to the lattice plane responsible for the HOLZ line. An incident beam originating at the imaginary source \( S \) and parallel to the direction \( SP \) fulfills the Bragg condition for a crystallographic plane. By diffraction, a part of the intensity of the primary beam is transferred to the excess beam. The defect beam is visible in the diffraction plane at the point \( P \). In a three-dimensional scheme, \( P \) is the point of the HOLZ line closest to the center \( O \) of the primary disk.

To determine the position \( P \) of the line in the pattern, it is necessary to calculate the angle \( \varphi \) and to use the length \( L \). The angle \( \theta \) for scattering of a lattice plane \( \vec{h} \) is given by the Bragg condition

\[
\theta = \arcsin \left( \frac{1}{2} \sqrt{\vec{h}^T G^{-1} \vec{h}} \right)
\]  

(6.1)

\( G \) is the metric tensor defined by \([54]\)

\[
G = \begin{pmatrix}
a^2 & ab \cos \gamma & ac \cos \beta \\
ab \cos \gamma & b^2 & bc \cos \alpha \\
ac \cos \beta & bc \cos \alpha & c^2
\end{pmatrix}
\]  

(6.2)

where \( a, b, c, \alpha, \beta, \) and \( \gamma \) are the lattice parameters of the crystal. The angle \( \Psi \) between the lattice plane \( \vec{g} \) and the zone axis \( \vec{n} \) may be calculated using the scalar product:

\[
\Psi = \arcsin \left( \frac{\vec{n} \cdot \vec{h}}{\sqrt{(\vec{n}^T G \vec{n})(\vec{h}^T G^{-1} \vec{h})}} \right)
\]  

(6.3)
Using eq. 6.1 and eq. 6.3, we have

$$\varphi = \Psi - \theta, \quad d = L \tan \varphi$$  \hspace{1cm} (6.4)

where $d$ is the distance from the center $O$ of the pattern to the point $P$ on the screen and $L$ is the camera length of the microscope (the distance $|OS|$ in Fig. 6.1). The

![Figure 6.1: Schematic diagram of excess and defect line formation for HOLZ reflections.](image)

relative direction of the lines in the pattern can be described using angles between them. These are the projections of the angles between the reflecting planes on the screen, perpendicular to the zone axis. To calculate the angle between two planes $h_1$ and $h_2$ one determines first the projections $h_1'$ and $h_2'$ of their normals:

$$h_{1,2}' = G^{-1}h_{1,2} - \frac{\hat{n} \cdot h_{1,2}}{\hat{n}^T G\hat{n}}$$  \hspace{1cm} (6.5)

The angle between two HOLZ lines, defined as the angle $\alpha$ between $h_1'$ and $h_2'$, is given by

$$\cos \alpha = \frac{h_1'^T G h_2'}{\sqrt{(h_1'^T G h_1')(h_2'^T G h_2')}}$$  \hspace{1cm} (6.6)

### 6.2 Dynamical corrections

The kinematical theory predicts exactly the position of a HOLZ line for the two-beam case, in which all other beams have zero intensity. Multiple diffraction between incident and diffracted beam only changes the intensity of the HOLZ line, but not its
position. In reality, there are always other weak beams present, and the kinematical approach for the determination of the HOLZ line position fails. A "dynamical HOLZ line shift" occurs owing to interactions with mainly ZOLZ reflections. The best way to quantify the shift is by many-beam dynamical calculation as described in chapter 4. However, such calculations are not practical for the routine analysis of HOLZ line patterns, since they need extensive computation. One way to approximately include the many-beams effect is a perturbation method. If there is a strong coupling in the ZOLZ and a weak interaction with a HOLZ reflection, the latter may be treated as a perturbation [53, 54]. The dispersion surface [57] is calculated from the unperturbed scattering matrix (see eq. 4.8) containing the potential of the ZOLZ reflections. Perturbation theory indicates that the dynamical shift depends only on the eigenvalues $\kappa_i$ of the scattering matrix. The deviation parameter $s_g$ is modified to $s_g - \kappa_i$, and the position of the HOLZ line $\bar{g}$ follows from the condition $s_g - \kappa_i = 0$ [54].

In the kinematical case, $k_\% = 0$ is assumed. The important interaction is that between the HOLZ reflection and the dispersion surface of the ZOLZ closest to the sphere of radius $k$ drawn about the origin. This follows because the smaller $\kappa$, the larger is the excitation of the beam. For crystal orientations without strong scattering conditions, branch 1 of the dispersion surface is nearest to the $k$ sphere and is responsible for the shift. The HOLZ line position in the central disc can be approximated by finding the intersection between the branch 1 and a $k$ sphere centred on the HOLZ reflection as shown in Fig. 6.2 (that is equivalent to the condition $s_g - \kappa_1 = 0$). In Fig. 6.2 the incident and diffracted beam starting in $Q$ have the correct dynamical trajectories. If the shift is not considered, the kinematic approach yields the wrong intersection point $P$, for which incident and diffracted beam have the same wavelength. We may assume that, over a small region of the dispersion surface, $\kappa$ is approximately constant. Then the effects of dynamical dispersion may be thought of as a correction to the accelerating voltage, and accommodated by a new wave vector $k'$ with $k' = k - \Delta k$ [52]. A kinematic simulation using the new wave vector $k'$ will give then the correct dynamical shift. Since branch 1 is relatively flat at the zone center, the correction may be assumed as nearly orientation-independent over the central disk. However, it differs from zone to zone, and it is lowest for the highest HOLZ layer. Referring to Fig. 6.2 we have the following relationship:

$$\sqrt{k^2 - (k + \kappa_1 - H)^2} = \sqrt{g^2 - H^2}$$ (6.7)

Furthermore,

$$\frac{H}{g} = \frac{g}{2k'}$$ and thus $g^2 = 2Hk'$ (6.8)
Figure 6.2:
Combined diagram for kinematic and dynamical diffraction into a HOLZ reflection $\vec{g}$. The construction with incident and diffracted beam intersecting in P relates to kinematic scattering with incident wave vector $\vec{k}$. The construction in Q shows the correct dynamical trajectories. Using the corrected vector $\vec{k}'$ the kinematic approach yields a correct position for the defect line beam parallel to QO.
Inserting eq. 6.8 in eq. 6.7 yields

\[ \Delta k = k - k' = \frac{\kappa_1 k}{H} + \frac{\kappa_1^2}{2H} - \kappa_1 \]

(6.9)

In the small-angle approximation we have \( \Delta k \approx \kappa_1 k / H \). The relativistic wave vector is given by

\[ k = \frac{1}{\hbar} \sqrt{2e} + \frac{m_0c^2}{e} \left( 1 + \frac{eU}{2m_0c^2} \right) \]

(6.10)

where \( c \) is the velocity of light and \( m_0 \) the rest mass of the electron.

One finds then the correction voltage as a function of \( \Delta k \):

\[ \Delta U = - \left( U + \frac{m_0c^2}{e} \right) + \sqrt{\left( U + \frac{m_0c^2}{e} \right)^2 + \left( \frac{\hbar c}{e} \right)^2 (\Delta k^2 + 2k\Delta k)} \]

(6.11)

The corrected voltage \( U' = U + \Delta U \) is lower than the nominal one.

As explained before, \( \kappa_1 \) of the first branch of the dispersion surface may be calculated solving the eigenvalue problem for the unperturbed scattering matrix. However, since only \( \kappa_1 \) is required for the purpose of HOLZ line simulation, it would clearly be advantageous to have a more rapid method of estimating \( \kappa_1 \). The following approximation based on perturbation theory has been proposed by Bird [55]:

\[ \kappa_1 = - \left( \frac{2me}{h^2} \right)^2 \frac{1}{2k} \sum_{\vec{g} \neq 0} \frac{|g^2|^2}{|g|^2} \]

(6.12)

The summation is only over reflection \( \vec{g} \) in the ZOLZ. The dynamical correction is thus lowest for high-index zone axes, or may be greatly reduced by using zone axes where \( \kappa_1 \) is small.

### 6.3 Uniqueness in the evaluation of lattice parameters

The uniqueness in the lattice parameter determination from experimental HOLZ patterns is still an open question. Several authors have tried to solve the problem in different ways or to discuss the limitations imposed by the experimental conditions. Maier et al. [67] analyzed the general problem of the unique evaluation of HOLZ lines. Within the experimental error it is possible to find more than one set of parameters (lattice parameters, camera length) that fit the given HOLZ pattern. To avoid this multiplicity, the authors suggest either to reduce the number of parameters to fit or to take several patterns with different orientations from the same area. Considerations about the sample, such as surface relaxation or strain due to particular conditions, permit to limit the parameters to be fitted, keeping the others constant,
i.e. using the values of the unstrained crystal. Two main methods have been used to compare the experimental data with the theoretical ones. Rozeveld and Howe [66] considered the ratios between different areas delimited by the HOLZ lines. Zuo [43] characterized the patterns determining distances between line intersections. Both procedures allow the lines to be evaluated without knowing the exact orientation of the crystal. In either method a correction voltage for the kinematic simulation was used. Deininger et al. [68] applied many-beam dynamical theory to simulate the CBED patterns and to compare them with experimental ones. The method requires zero-loss filtered images to compare also the intensities of the reflections. Here, the position of the lines can be better predicted than in the methods relying on voltage correction and kinematical simulations. Furthermore the comparison of line scan profiles in the CBED patterns permits structure factors to be determined. Recently Wittmann et al. [69] employed the algorithm developed by Rozeveld and Howe [66] to determine all six parameters of a triclinic distorted structure. Within the experimental accuracy, no unique solution was found.

The uniqueness of the parameter set can be proved only within an interval around the solution, using linearized forms of eqs. 6.4 and 6.6. Consider small deviations \( \Delta a, \Delta b, \Delta c \) around the cubic lattice constant \( a_0 \), and \( \alpha, \beta, \gamma \) close to 90\(^\circ\). The term \( \vec{n}^T G \vec{n} \), with \( \vec{n} = [n_1, n_2, n_3] \), is written explicitly

\[
\vec{n}^T G \vec{n} = n_1^2 a^2 + n_2^2 b^2 + n_3^2 c^2 + 2(n_1 n_2 a b \cos \gamma + n_1 n_3 a \cos \beta + n_2 n_3 b \cos \alpha) \quad (6.13)
\]

Substituting \( a = a_0 + \Delta a, b = a_0 + \Delta b, c = a_0 + \Delta c \) in eq. 6.13 and eliminating the quadratic terms we obtain

\[
\vec{n}^T G \vec{n} \approx a_0^2 \vec{n}^2 + 2a_0(n_1^2 \Delta a + n_2^2 \Delta b + n_3^2 \Delta c) + 2a_0^2(n_1 n_2 a b \cos \gamma + n_1 n_3 a \cos \beta + n_2 n_3 b \cos \alpha) \quad (6.14)
\]

Using the binomial approximation \((1 + x)^m \approx 1 \pm mx\) we have

\[
\frac{1}{\sqrt{\vec{n}^T G \vec{n}}} \approx \frac{1}{a_0 |\vec{n}|} \left( 1 - \frac{(n_1^2 \Delta a + n_2^2 \Delta b + n_3^2 \Delta c)/a_0 + n_1 n_2 a b \cos \gamma + n_1 n_3 a \cos \beta + n_2 n_3 b \cos \alpha)}{|\vec{n}|^2} \right) \quad (6.15)
\]

In a similar way, for the reflection \( \vec{h} = (h_1, h_2, h_3) \), neglecting quadratic cosine terms,

\[
\vec{h}^T G^{-1} \vec{h} \approx \frac{h_1^2}{a^2} + \frac{h_2^2}{b^2} + \frac{h_3^2}{c^2} - 2\frac{h_1 h_2}{ab} \cos \gamma - 2\frac{h_1 h_3}{ac} \cos \beta - 2\frac{h_2 h_3}{bc} \cos \alpha \quad (6.16)
\]

and it follows

\[
\frac{1}{\sqrt{\vec{h}^T G \vec{h}}} \approx \frac{1}{|\vec{h}|} \left( 1 + \frac{(h_1^2 \Delta a + h_2^2 \Delta b + h_3^2 \Delta c)/a_0 + h_1 h_2 \cos \gamma + h_1 h_3 \cos \beta + h_2 h_3 \cos \alpha)}{|\vec{h}|^2} \right) \quad (6.17)
\]
Thus the linearized form of eq. 6.4 for the distance \( d \) of the HOLZ line \( \vec{n} \) from the exact pole \( \vec{n} \) is

\[
\left( \frac{|\vec{n}|^2 - U|\vec{n}|^2 + T|\vec{n}|^2}{|\vec{n}|^3} \right) \frac{\vec{\lambda}}{|\vec{n}|} \approx d
\]

(6.18)

where

\[
U = \frac{n_1^2 \Delta a + n_2^2 \Delta b + n_3^2 \Delta c}{a_0} + n_1 n_2 \cos \gamma + n_1 n_3 \cos \beta + n_2 n_3 \cos \alpha
\]

(6.19)

\[
T = \frac{(h_1^2 \Delta a + h_2^2 \Delta b + h_3^2 \Delta c)}{a_0} + h_1 h_2 \cos \gamma + h_1 h_3 \cos \beta + h_2 h_3 \cos \alpha
\]

(6.20)

The analogon of eq. 6.4 in the cubic case, for small scattering angles, is

\[
d = L \left( \frac{\vec{n} \cdot \vec{h}}{|\vec{n}|} - \frac{\lambda}{2a_0 |\vec{n}|} \right)
\]

(6.21)

Comparison of eqs. 6.18 and 6.21 yields

\[
\frac{U}{|\vec{n}|^2} \approx \left( \frac{1}{|\vec{h}|^2} + \frac{\lambda}{2a_0 |\vec{n}| \cdot |\vec{h}|} \right) T
\]

(6.22)

In eq. 6.22 we assume that the real set of lattice parameters is that of the cubic case. Thus, the correct solution of the problem is the trivial case \( \Delta a = \Delta b = \Delta c = 0 \), \( \alpha = \beta = \gamma = 90^\circ \). The left term of eq. 6.22 depends on the crystal orientation and not on the individual reflections. For every HOLZ line the term on the right changes according to the different \( (h_1, h_2, h_3) \). To find the set of variables \( \Delta a, \Delta b, \Delta c, \alpha, \beta, \gamma \) fulfilling eq. 6.22, one considers the system of equations given by 6 different HOLZ lines. The system may be written in the form

\[
M \vec{X} = \vec{0}
\]

with

\[
M_{ij} = H_i^j \left( \frac{1}{|\vec{h}_i|^2} + \frac{\lambda}{2a_0 |\vec{n}| \cdot |\vec{h}_i|} \right) - \frac{N_j}{|\vec{n}|^2}
\]

(6.23)

and with six-dimensional vectors

\[
\vec{N} = (n_1^2, n_2^2, n_3^2, n_2 n_3, n_1 n_3, n_1 n_2)
\]

(6.24)

\[
\vec{H}_i^j = ((h_1^2)^j, (h_2^2)^j, (h_3^2)^j, h_1 h_2^i h_3^j, h_1 h_3^i h_2^j, h_2 h_3^i h_1^j)
\]

(6.25)

\[
\vec{X} = (\Delta a/a_0, \Delta b/a_0, \Delta c/a_0, \cos \alpha, \cos \beta, \cos \gamma)
\]

(6.26)

The condition for eq. 6.23 to have only a trivial solution is \( \det(M) \neq 0 \), which is usually fulfilled.

For symmetrical zone axes and reducing the free parameters, it is possible to solve the problem without the linearization on a given point. Consider the case with \( \vec{n} = [111] \), \( a = b = c \), and \( \alpha = \beta = \gamma \). We assume again the correct set of lattice parameters
to be those of the cubic structure, i.e. the distance from a line to the center is given by eq. 6.21. Comparing eq. 6.4 with 6.21 and solving for the parameter $a$ yields

$$a = \frac{\lambda}{2r} \left( \frac{\vec{h}_1 h_1 + h_2 + h_3}{2a_0} - \frac{\vec{h}_1 + h_2 + h_3}{\sqrt{3}|\vec{h}|} \right)^{-1}$$

where

$$r = \sqrt{\frac{|\vec{h}|^2 + (|\vec{h}|^2 - 2(h_1 h_2 + h_3 h_3 + h_2 h_3) \cos \alpha)}{\cos \alpha - \cos(2\alpha)}}$$

Eq. 6.27 yields, for a given $\alpha$, the value of $a (= b = c)$, so that the distance $d$ from the center of the considered line $\vec{h}$ remains the same as for the cubic case. The possible solutions for one line may be represented by a curve in the two-dimensional space of $a$ and $\alpha$ (see Fig. 6.3). If two reflections are considered, the solution is given by the intersection of the two curves. Figure 6.3 shows the function 6.27 for different reflections of Si(111) and 100 kV acceleration voltage. The curves intersect at $a = 5.43 \text{ Å}$, the lattice constant of Si, and $\alpha = 90^\circ$. Figure 6.4 shows the curves over a wide angular range. For reflections leading to a very similar slope, the determination of the intersection point is accordingly more difficult. This implies that a change of the lattice parameters according to one curve $a(\alpha)$ does not significantly influence the HOLZ line pattern. The positions of the HOLZ lines are mathematically different for different lattice parameters $a$, $\alpha$, but experimentally the HOLZ line patterns are undistinguishable. The choice of the HOLZ lines may influence the accuracy of the
lattice parameter determination, especially when experimental errors are present. Over the displayed range there is only one intersection point for the three curves, indicating that the solution for the considered case is unique. The parameter $a$ for small angles $\alpha$ diverges to large values, and coincidence points for the three reflections are no longer present.

The general problem of the lattice parameter determination from HOLZ patterns is more complicated than discussed above. Usually the exact orientation of the crystal is not known: the distance of the HOLZ line from the center cannot be determined, and only distances between the HOLZ-line intersections are available. Therefore the zone axis $\vec{n}$ has to be treated as a further variable. The vector $\vec{n}$ in eq. 6.18 may be substituted by $\vec{n}/|\vec{n}| + \Delta \vec{n}$, with $\Delta \vec{n} = [\Delta n_1, \Delta n_2, \Delta n_3]$. To simplify the equation we may put the condition $|\vec{n}|/|\vec{n}| + \Delta \vec{n}| = 1$, or, neglecting quadratic terms in $\Delta \vec{n}$,

$$ n_1 \Delta n_1 + n_2 \Delta n_2 + n_3 \Delta n_3 = 0 \quad (6.29) $$

On the other hand, to fully characterize the HOLZ pattern, the angle between the lines may also be considered. In a similar way as for eq. 6.4, eq. 6.6 can be approximated by a linear form on a restricted interval. The angle $\delta_{lm}$ between two HOLZ lines $\vec{h}^l$ and $\vec{h}^m$ is given by (see eqs. 6.5 and 6.6)

$$ \cos \delta_{lm} = \frac{\left(\frac{\vec{h}^l}{G} \vec{h}^m \right) - \frac{(\vec{n} \cdot \vec{h}^l)(\vec{n} \cdot \vec{h}^m)}{\vec{n}^2 G \vec{n}^2}}{\sqrt{\left(\frac{\vec{h}^l}{G} \vec{h}^l \right) \left(\frac{\vec{h}^m}{G} \vec{h}^m \right) - \frac{(\vec{n} \cdot \vec{h}^l)^2}{\vec{n}^2 G \vec{n}^2}} \sqrt{\left(\frac{\vec{h}^m}{G} \vec{h}^m \right) \left(\frac{\vec{h}^l}{G} \vec{h}^l \right) - \frac{(\vec{n} \cdot \vec{h}^m)^2}{\vec{n}^2 G \vec{n}^2}}} \quad (6.30) $$

where $\vec{n}$ is the zone axis, and $G$ is the metric tensor for the considered crystal. In a linearized form, with small deviations $\Delta a$, $\Delta b$, $\Delta c$ around the cubic lattice constant
\(a_0\), and \(\alpha, \beta, \gamma\) close to 90°, eq. 6.30 may be approximated neglecting quadratic terms and using the binomial approximation. The expression \(\frac{1}{n T G} n\) may be calculated using eq. 6.14:

\[
\frac{1}{n T G} n \approx \frac{1}{a_0^2 n^2} \left(1 - \frac{2}{|n|^2} \sum_{i=1}^{3} \sum_{j \geq i} A_{ij} n_i n_j\right)
\]

where

\[
A = \begin{pmatrix}
\Delta a \\
\cos \gamma \\
\cos \beta \\
\cos \alpha
\end{pmatrix}
\]

(6.32)

From eq. 6.16 one obtains

\[
(h^i)^T G^{-1} h^i \approx \frac{1}{a_0^2} \left(\frac{|h^i|^2}{2J^2} - 2 \sum_{j=1}^{3} \sum_{k \geq j} A_{jk} h_j^i h_k^i\right) i = 1, m
\]

(6.33)

Similar considerations yield

\[
(h^i)^T G^{-1} h^m \approx \frac{1}{a_0^2} \left(\sum_{k=1}^{3} (1 - A_{kk}) h_k^i h_k^m - \sum_{j=1}^{3} \sum_{i=1}^{3} A_{ij} h_j^i h_j^m\right)
\]

(6.34)

Applying the binomial approximation gives

\[
\frac{1}{\sqrt{(h^i)^T G^{-1} h^i - \frac{|h^i|^2}{|n|^2}}} \approx \sqrt{C_i} \left(1 - \frac{1}{2C_i} \left(\frac{(S^i)^2}{|n|^2} - \sum_{j=1}^{3} \sum_{k \geq j} A_{jk} h_j^i h_j^i\right)\right), i = 1, m
\]

(6.35)

where

\[
S^i = \bar{n} \cdot h^i, \quad C_i = \sqrt{|h^i|^2 - \frac{(S^i)^2}{|n|^2}}
\]

(6.37)

Inserting eqs. 6.34 and 6.36 into eq. 6.30 and neglecting quadratic terms, the angle between two HOLZ lines can be approximated by

\[
\sum_{i=1}^{3} \sum_{j \geq i} A_{ij} \left(\frac{2n_i n_j}{|n|^4} - h_i^j h^i - h_i^m h^m + \left(h^i \cdot h^m - \frac{S^i S^m}{|n|^2}\right) \sum_{k=1}^{3} \sum_{l=1}^{3} \sum_{m=1}^{3} \frac{1}{C_k} \left(h_i^k h_j^k - (S^k)^n n_i n_j\right)\right)
\]

\[
+ \frac{1}{C_l C_m} \left(h^i \cdot h^m - \frac{S^i S^m}{|n|^2}\right) \approx \cos \delta_{lm}
\]

(6.38)

The second line of eq. 6.38 gives the exact angle for the cubic case. The first line gives the variation due to deviations from the cubic symmetry. In eq. 6.38, the term \(\Delta \bar{n}\) may be neglected, because the angle between the lines, and in general the whole pattern, does not essentially change if the crystal is slightly tilted. Combining
eqs. 6.18, 6.29, and 6.38 the problem is fully determined in a reasonably small interval around the solution of the unstrained crystal. With six HOLZ lines, which are commonly used in our simulations, twelve independent equations are available, six for the distances from the center (eq. 6.18), five for the angles between the lines (eq. 6.38), and one for the norm of $\vec{n}/||\vec{n}|| + \Delta \vec{n}$. This system of equations is sufficient to determine the six lattice parameters, the three components of the zone axis, and the camera length $L$.

These theoretical considerations suggest that it is possible to determine a unique set of lattice parameters from a HOLZ pattern. As we will see in the next sections, the theoretical predictions are unfortunately not applicable to the experimental case. Measurement errors affect the accuracy of the evaluation and limit the efficiency of the method.

6.4 Hough transformation

In experimental patterns, the lines are often blurred owing to inelastic scattering. For strong reflections, the lines become curved close to the intersection point, where they interact with each other resulting in deviations from the approximate straight trajectories. In some cases, crystal defects can also affect a part of a line, changing its intensity or its sharpness. On the other hand, the kinematic simulations gives the exact line position for Bragg condition, i.e. an exactly located sharp line. To compare simulations and experimental results, it is necessary to reduce the experimental lines to perfect lines which can be described analytically. Some averaging of the experimental positions, depending on the intensities has to be done. In the Hough transformation every line present in the original image (or real space) is transformed into a point in the so-called Hough space [70]. A line in the two-dimensional space of the diffraction pattern always can be described by two parameters. If we choose the angle $\beta$ between the line $l$ and the abscissa, and $r$ the vector perpendicular to $l$ (see Fig. 6.5), then the line is described by the equation

$$x\cos\beta + y\sin\beta = |\vec{r}|$$

(6.39)

where $x$ and $y$ are cartesian coordinates in the diffraction pattern. The Hough transformation of line $l$ described by $r$ and $\beta$ is a point with coordinates $r$ and $\beta$ in Hough space (see Fig. 6.6). The transformation $H(r, \beta)$ is given by

$$H(r, \beta) = \sum_{(x,y) \in l} I(x, y)$$

(6.40)

where $I$ is the value of the intensity at coordinates $(x, y)$ in the experimental pattern. The choice of the coordinate system is arbitrary, because only the relative position
Figure 6.5:

HOLZ line pattern with analytical description of one line.

Figure 6.6:

Hough transformation of the pattern in Fig. 6.5. The small squares with a white spot are the regions for the determination of the spot center.
between the lines is important. The summation is discrete over a finite number of positions in the experimental image usually recorded with a CCD camera or digitized from a photograph. The result of the transformation in Hough space is an image with several spots of varying intensity and size as shown in Fig. 6.6. Every spot indicates a HOLZ line and its center yields the parameters $r$ and $\beta$. To find the spot center, the contrast of the image in a small square around the spot is enhanced by a digital filter and a center of mass relating to the intensity is determined. A Hough space containing now only the spot centers is transformed back to real space and a digitized image with reconstructed lines is obtained. Figure 6.7 shows the original image overlapped by the lines selected in Fig. 6.6 and transformed back.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure6.7.png}
\caption{Original image overlapped with some reconstructed lines.}
\end{figure}

### 6.5 Least-squares fitting of the lattice parameters

A direct way to determine lattice parameters through an analytic solution involving the eqs. 6.4 and 6.6 is too complicated. An alternative is to find the parameters by comparing a simulated HOLZ pattern with an experimental one. The comparison is achieved using a best-fit parameter $\chi$ defined by

$$
\chi^2 = \sum_i \frac{(d_i^{\text{theory}} - d_i^{\text{exp}})^2}{\sigma_i^2}
$$

(6.41)
where \(d_i\) are the distances between the line intersections, and \(\sigma^2\) their variances. Using these distances, a knowledge of the exact orientation of the crystal with respect to the incident beam, i.e. the exact zone axis, is not necessary. The relative positions of the lines in a pattern change only in a negligible way for small variations of the zone axis. A slight change of orientation displaces the pattern as a whole on the screen, without influencing the relative distances between the intersection points.

The coordinates \(x\) and \(y\) for the intersection point of two lines \(p\) and \(q\) are given by

\[
x = \frac{q_2p_1^2 - p_2q_1^2}{p_1q_2 - q_1p_2}, \quad y = \frac{p_1q_2^2 - q_1p_2^2}{p_1q_2 - q_1p_2}
\]

\((6.42)\)

The vectors \(\vec{p} = (p_1, p_2)\) and \(\vec{q} = (q_1, q_2)\) are perpendicular to the respective HOLZ line and point from the center to the corresponding line. Their lengths can be calculated using eq. 6.4 and their directions with eq. 6.6. The distances between the line intersections on the experimental image are evaluated by means of a Hough transformation. After the lines are digitized and converted into an analytical formula, eq. 6.42 can be applied to find the intersection points. The refinement of lattice parameters and camera length is achieved by a minimization of the function \(\chi^2\). The "dfpmin" algorithm implemented in the computing environment IDL\textsuperscript{®} and based on the Broyden-Fletcher-Goldfarb-Shanno variant of the Davidon-Fletcher-Powell method is employed (see section 10.7 in [64]).

To test the least-squares fitting method, for a given set of lattice parameters, wavelength, and camera length, distances between the line intersections were calculated and inserted in the \(\chi^2\) function instead of the experimental values. With a fixed camera length the algorithm converges to the same minimum of \(\chi\) for different starting lattice parameters and gives a solution with a relative error in the lattice parameters of \(10^{-8}\) and \(\chi^2 = 10^{-22}\). If the camera length is treated as a fitting parameter, the algorithm has some difficulties to converge and the final result depends on the starting point. The originally selected set of parameters can not be reached. Therefore it is better to use a constant camera length in the algorithm and to start the fitting procedure several times with different values of the camera length, which gives results comparable to those obtained using a known camera length. The camera length is the critical factor for the convergence of the algorithm, because it scales the distances in the pattern and may be approximately compensated by a scaling of \(a, b,\) and \(c\). With the ratio of the distances between the line intersections the camera length would be eliminated in the calculation, but the same kind of scaling remain. Refinements with the ratio of the triangular areas delimited by the HOLZ lines, or with the ratio of the distances were also examined, but similar problems
in the convergence of the algorithm were encountered. Figure 6.8 shows lines of equal magnitude of \( \log(\chi^2) \) on the \((\Delta a, \cos \alpha)\) plane assuming values for Si[111] at 100 kV, \( \Delta a = \Delta b = \Delta c \) and \( \alpha = \beta = \gamma \).

Lines of equal magnitude of \( \log(\chi^2) \) using calculated values for Si[111] at 100 kV, \( \Delta a = \Delta b = \Delta c \) and \( \alpha = \beta = \gamma \). The \( \chi^2 \) function has a very flat, elongated minimum region, where the algorithm is not very stable, and the values are close to the double precision limits of the computing machine. The differences in the \( \chi^2 \) function near the minimum are very small, i.e. for specific large variations of the lattice parameters, the position of HOLZ lines in the pattern do not vary very much. Figures 6.9 and 6.10 show two kinematic simulations of Si[111] at 100 kV with two different sets of lattice parameters in the region around the minimum of \( \chi \). The difference of the two patterns is almost imperceptible, although the lattice parameters differ considerably. This fact makes it difficult to analyze experimental patterns. Even under optimal conditions, the accuracy of the measurements is limited by the width of the HOLZ lines depending on the sample thickness. The errors in the line positions lead to a large degree of uncertainty in the parameter sets. The best way to overcome this problem, as already mentioned above, consists in reducing the fit variables or evaluating HOLZ patterns from different orientations. Refinements
Figure 6.9:
Kinematic simulation of Si[111] at 100 kV with lattice parameters $a = b = c = 5.412 \text{ Å}$ and $\alpha = \beta = \gamma = 90^\circ$.

Figure 6.10:
Kinematic simulation of Si[111] at 100 kV with lattice parameters $a = b = c = 5.434 \text{ Å}$ and $\alpha = \beta = \gamma = 89.88^\circ$. 
with more than one orientation have been performed. The squares of the differences in each pattern are put in an unique summation, giving a total $\chi$ function for all the patterns. Using this approach, with information from different orientations, the algorithm is very stable, and converges to accurate lattice parameter values. The camera lengths of the different patterns are also fitted accurately in this procedure, irrespective of the starting values.

### 6.6 Voltage calibration

For a comparison of experimental and simulated HOLZ patterns it is necessary to know exactly the electron wavelength used to take the experimental images. The nominal voltage indicated by the microscope is in general not very accurate. The wavelength is determined through a calibration using a Si standard. Figure 6.11 shows a HOLZ pattern of pure Si along the [113] zone axis taken at 230 kV. In this case a refinement of the wave length and the camera length, keeping a fixed lattice constant for Si ($a_0 = 5.4309 \text{ Å} [59]$) is performed. The voltage found for Fig. 6.11 is $(229.0 \pm 0.1) \text{ kV}$. Figure 6.12 shows a kinematic simulation of Si[113] at 229 kV. The relative positions of the line intersections coincide with the experimental pattern. In this simulation no dynamical effects are taken into account. The correction

![HOLZ pattern of pure Si at 230 kV.](image)
Figure 6.12: Kinematic simulation of [113] HOLZ pattern at 229 kV.

Voltage for Si[113] at 230 kV can be calculated using eqs. 6.12, 6.9 and 6.11. For the Fourier coefficients $V_g$ of the lattice potential the routine "vg2" of the standard EMS simulation package of Stadelmann [60] is used. The routine relies on the atomic form factors of Weickenmeier and Kohl [61]. The Debye-Waller factor $\exp(-Bg^2/2)$, influencing the crystal potential, is calculated using the expression [62]

$$B = \frac{11492}{A T_D^2} \psi(T_D/T) + \frac{2873}{AT_D}$$  \hspace{1cm} (6.43)

where $A$ is the atomic weight, $T_D$ is the Debye temperature, and $\psi(x) = \frac{1}{x} \int_0^x \frac{\xi}{e^{\xi} - 1} d\xi$. Assuming $T_D = 645$ K [63] for Si, one obtains a Debye parameter $B$ at 300 K of about 0.32 Å². The calculated correction is -1.47 kV. Thus the nominal voltage of 230 kV corresponds to a kinematically calibrated value of $(229 \pm 0.1)$ kV and a dynamically corrected voltage of $(230.5 \pm 0.1)$ kV. A dynamical simulation of Si[113] at 230.5 kV (Fig. 6.13) shows in fact that the intersections are in good agreement with Fig. 6.11.

6.7 Strain measurement in a Si grain

Strong distortions of the crystal due to dislocations or to thermal stresses hamper a good evaluation of HOLZ patterns. Blurring or splitting of lines and general loss of contrast are the most common problems. The size of the electron probe determines the quality of the image. Since every HOLZ line contains information from the whole
probed area, strong changes of the lattice parameters within this region cause a broadening of the lines with consequent losses in quality and accuracy. To overcome these problems, simulations with a finite probe size as for the LACBED case may be useful. This technique will be discussed in chapter 8.

Many CBED images were taken on different places for both reinforced materials. The best conditions for a good image quality were found for Si grains with thicknesses exceeding 100 nm. A sufficient thickness is necessary to have enough diffracted intensity. Furthermore, from eq. 5.1 it is clear that the HOLZ lines become sharper with increasing thickness. On the other hand, regions of excessive thickness are not ideal because of strong inelastic scattering, unless an energy filter is used. In Si, very few dislocations are present, and the HOLZ lines are therefore not as blurred as in patterns taken in Al grains. Nevertheless, close to the reinforcements, the thermal stresses are high enough to affect the sharpness of the HOLZ lines. Thus, the places available for a strain analysis are scarce. In Fig. 5.5 a large Si grain, satisfying the required conditions, is shown. CBED images in the positions A (Fig. 5.8) and B (Fig. 5.9) were taken with 230 kV nominal voltage, at liquid nitrogen temperature, and were digitized. In Fig. 6.14 digitized lines, belonging to Fig. 5.8, are shown. The [001] direction of the Si grain is approximately parallel to the foil normal. If only a
Figure 6.14:
Digitized HOLZ lines of Fig. 5.8. The pattern is rotated for a better comparison with the corresponding kinematic simulation (see Fig. 6.16).

planar distortion is assumed, which is not improbable in a thin TEM foil, we may choose the refinement parameters $a$, $b$, $\gamma$, and the camera length $L$. The intersection distances of the following six lines are used: $(1 1 1 3)$, $(1 1 3 3)$, $(3 1 1 5)$, $(3 3 1 5)$, $(3 1 1 3)$, $(1 1 3 3)$. An acceleration voltage of 229 kV is employed for the kinematical simulation, according to the above discussion. For position A the following values for the minimal $\chi^2$ ($= 1.56$) were found: $a = 5.429$ Å, $b = 5.436$ Å, $\gamma = 90.3^\circ$, $L = 1262$ mm. In position B the following values were obtained: $a = 5.415$ Å, $b = 5.442$ Å, $\gamma = 90.4^\circ$, $L = 1244$ mm. In this case the minimal $\chi^2$ ($= 7.03$) is much worse than in A. In the given $\chi^2$ values, the variances $\sigma_i^2$ were not considered and all summation terms were equally weighted. From the minimum $\chi_{\text{min}}$ a brief estimate of the average error in the distances may be done. If we assume that all measured distances $d_i^{\text{exp}}$ have the same error $\sigma$ ($\sigma_i = \sigma$), it follows that $\sigma^2 = \frac{\sum_i^N (d_i^{\text{the}}(\chi_{\text{min}}) - d_i^{\text{exp}})^2}{(N - M)}$ (eq. 15.1.6 in [64]), where $d_i^{\text{the}}(\chi_{\text{min}})$ are the theoretical distances obtained with the parameters of the best fit, $N$ is the number of measured distances, and $M$ is the number of fitting parameters. With $\chi^2 = 7.03$, $N = 105$, and $M = 4$ one obtains $\sigma = 0.2$ mm. The error $\sigma(a_j)$ associated with the estimated lattice parameter $a_j$
may be evaluated using

\[ \sigma^2(a_j) = \sum_{i=1}^{N} \sigma_i^2 \left( \frac{\partial a_i}{\partial d_i} \right)^2 \]  

(6.44)

For the latter case the relative error in the three parameters is about $10^{-3}$.

Measurements with the same imaging conditions in the same places at room temperature were also performed. In A the refinement gives: $a = 5.429 \, \text{Å}, b = 5.434 \, \text{Å}, \gamma = 89.3^\circ, \chi^2 = 0.49$. In B: $a = 5.421 \, \text{Å}, b = 5.438 \, \text{Å}, \gamma = 90.3^\circ, \chi^2 = 4.15$. The error for position B, with the worst $\chi$, is about $10^{-3}$.

These quite large errors suggest that the assumption about planar strain made above does not correspond to the real conditions. Therefore, refinements considering simultaneously several patterns taken at room temperature with the following orientations were performed: [012], [113], [129], [125]. In the $\chi$ function a variance $\sigma_i^2$ proportional to $(1 + d_i^{\text{exp}})^2$ was used. The parameters obtained in position A are: $a = 5.4452 \, \text{Å}, b = 5.4541 \, \text{Å}, c = 5.4303 \, \text{Å}, \alpha = 89.752^\circ, \beta = 89.563^\circ, \gamma = 89.868^\circ$, and $\chi^2 = 0.038$. In the position B: $a = 5.446 \, \text{Å}, b = 5.455 \, \text{Å}, c = 5.424 \, \text{Å}, \alpha = 89.55^\circ, \beta = 89.79^\circ, \gamma = 89.90^\circ$, and $\chi^2 = 0.32$. Figure 6.15 shows a HOLZ line pattern taken in A along the [125] zone axis. Kinematic simulations using the

![Image](image.png)

Figure 6.15:

HOLZ pattern at the position A of Si[125] at 230 kV nominal voltage.

refined parameters for the [113] and [125] zone axis in the position A are shown
in Figs. 6.16 and 6.17. Experimental and simulated patterns agree well, and the

Figure 6.16:

Kinematic simulation with refined parameters for Si[113] in the position A.

relative error in the lattice parameters estimated with eq. 6.44 is about \(2 \times 10^{-4}\) for \(\chi^2 = 0.038\). The use of more than one orientation improves the quality of the refinement. Several refinements with two, three and four orientations were performed, and in general the more orientations are used, the better is the fit. The difference of the minimal \(\chi^2\) values in A and B may be due to the bad reproducibility of the measurement. In fact, for each HOLZ pattern, the probed position on the sample has to be slightly changed, because of contamination. This may affect the compatibility between the different orientations and, depending on the place in the sample, give rise to considerable errors. The strain in the investigated positions A and B, may be evaluated using \(\frac{\Delta a}{a_0} \approx \varepsilon_{11}, \frac{\Delta b}{b_0} \approx \varepsilon_{22}, \frac{\Delta c}{c_0} \approx \varepsilon_{33}, \Delta\alpha \approx -2\varepsilon_{23}, \Delta\beta \approx -2\varepsilon_{13},\) and \(\Delta\gamma \approx -2\varepsilon_{12}\). Table 6.1 shows the evaluated strains in A and B. The directions in the grain corresponding to a and b are indicated in Fig. 5.5. Although the two positions are not close to each other, the strains are comparable, only the sign of \(\varepsilon_{33}\) reverses. The strains \(\varepsilon_{11}\) and \(\varepsilon_{22}\) are positive, in contradiction to an expected compressive thermal strain. The values of \(\varepsilon_{33}\) are relatively small, according to the assumption made in the refinement with only one orientation. The shear strains \(\varepsilon_{23}\) and \(\varepsilon_{13}\) are, on the contrary, not negligible. This explains the bad agreement between simulations and experimental patterns obtained in the fitting procedure using one
orientation. It is difficult to find a relation for the strain change between the two
different places. A simple explanation based on the distance from the fiber may not
be applied, because the grain is surrounded by several other fibers, and their stress
fields overlap. The strain in the middle of the grain is actually expected to be more
or less constant, being surrounded from all directions by fibers, and far away from
the strong influence of one particular fiber. The difference in the strains may be due
to the vicinity of a grain boundary or to other factors not easily predictable. Differ¬
ences between measurements at room temperature and liquid nitrogen temperature
are also difficult to interpret. The variations in the lattice parameters do not show
any clear tendency. As already mentioned above, the LACBED technique is suitable
to investigate the influence of the reinforcement, allowing the matrix closer to the
fibers to be analyzed. This topic will be discussed in chapter 8.

Table 6.1:
Table of the strains in the position A and B.

<table>
<thead>
<tr>
<th>×10^{-3}</th>
<th>( \varepsilon_{11} )</th>
<th>( \varepsilon_{22} )</th>
<th>( \varepsilon_{33} )</th>
<th>( \varepsilon_{23} )</th>
<th>( \varepsilon_{13} )</th>
<th>( \varepsilon_{12} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>2.8</td>
<td>4.4</td>
<td>0.18</td>
<td>2.2</td>
<td>3.8</td>
<td>1.1</td>
</tr>
<tr>
<td>B</td>
<td>2.9</td>
<td>4.6</td>
<td>-1.1</td>
<td>3.9</td>
<td>1.8</td>
<td>0.87</td>
</tr>
</tbody>
</table>
7 Elastic strains in the matrix

Since metals generally have larger thermal expansion coefficients than ceramics, and since the fabrication of MMCs involves solidification at a relatively high temperature, they often contain significant differential thermal contraction stresses after cooling to room temperature. The distribution of these thermal stresses can be predicted, for simple reinforcement-matrix configurations, by the elastic theory. In this chapter, for the formulation of the internal strains in the fiber-reinforced composite, we assume the fiber to be an isolated cylinder in an infinite matrix, without taking into account interactions between different fibers. Since the strain investigations by TEM are performed on thin samples, the problem of the surface relaxation will be also discussed.

7.1 Theory of elasticity

Starting point for the formalism of the theory of elasticity are the equations of equilibrium, the stress-strain relations and the strain-displacement relations. The equations of equilibrium may be written as [71]

\[ \text{Div} \sigma + \rho \ddot{f} = 0 \]  

where \( \rho \ddot{f} \) are the volume forces and \( \sigma \) is the stress tensor

\[ \sigma = \begin{pmatrix} \sigma_{11} & \sigma_{12} & \sigma_{13} \\ \sigma_{12} & \sigma_{22} & \sigma_{23} \\ \sigma_{13} & \sigma_{23} & \sigma_{33} \end{pmatrix} \]  

(7.2)

The strain tensor \( \varepsilon \) and the displacements \( u_i \) are related in the linear elastic theory through

\[ \varepsilon_{ij} = \frac{1}{2} (u_{i,j} + u_{j,i}) \]  

(7.3)

The index following the comma in eq. 7.3 indicates the partial derivative in curvilinear coordinates. Stress and strain in an isotropic body are related by the Hooke's law:

\[ \varepsilon_{11} = \frac{1}{E} (\sigma_{11} - \nu(\sigma_{22} + \sigma_{33})), \quad \varepsilon_{12} = \frac{1}{2G} \sigma_{12}, \]
\[ \varepsilon_{22} = \frac{1}{E} (\sigma_{22} - \nu(\sigma_{11} + \sigma_{33})), \quad \varepsilon_{13} = \frac{1}{2G} \sigma_{13}, \]
\[ \varepsilon_{33} = \frac{1}{E} (\sigma_{33} - \nu(\sigma_{11} + \sigma_{22})), \quad \varepsilon_{23} = \frac{1}{2G} \sigma_{23}, \]  

(7.4)

where \( E \) is Young's modulus, \( \nu \) is Poisson's ratio, and \( G = \frac{E}{2(1+\nu)} \) the shear modulus.
For an axially symmetrical stress distribution, the eqs. of equilibrium can be written in cylindrical coordinates:

\[
\begin{align*}
\frac{\partial \sigma_{rr}}{\partial r} + \frac{\partial \tau_{rz}}{\partial z} + \frac{1}{r} (\sigma_{rr} - \sigma_{\varphi\varphi}) + \rho f_r &= 0 \\
\frac{\partial \sigma_{rz}}{\partial r} + \frac{\partial \sigma_{zz}}{\partial z} + \frac{1}{r} \sigma_{rz} + \rho f_z &= 0 
\end{align*}
\] (7.5)

The relations of eq. 7.3 are written as

\[
\begin{align*}
\epsilon_{rr} &= \frac{\partial u_r}{\partial r}, & \epsilon_{\varphi\varphi} &= \frac{u_r}{r}, \\
\epsilon_{zz} &= \frac{\partial u_z}{\partial z}, & \epsilon_{rz} &= \frac{1}{2} \left( \frac{\partial u_r}{\partial z} + \frac{\partial u_z}{\partial r} \right) 
\end{align*}
\] (7.6)

### 7.2 Plane strains in a cylindrical system

The thermal strains of an infinitely-long cylinder immersed in an infinite matrix may be treated as plane strains. The eqs. 7.5 with no volume forces, in the plane case reduce to

\[
\frac{\partial \sigma_{rr}}{\partial r} + \frac{1}{r} (\sigma_{rr} - \sigma_{\varphi\varphi}) = 0 
\] (7.7)

Using eqs. 7.6, 7.4, and 7.7, considering only the plane components, one obtains

\[
\frac{d^2 u_r}{dr^2} + \frac{1}{r} \frac{du_r}{dr} - \frac{u_r}{r^2} = 0 
\] (7.8)

We assume that the radius of the matrix hole and of the cylinder at a temperature \( T_1 \) is \( R \). At a lower temperature \( T_2 \) the matrix hole tries to contract to \( r_m = R(1 + \alpha_m \Delta T) \) and the cylinder to \( r_c = R(1 + \alpha_c \Delta T) \), where \( \Delta T = T_2 - T_1 \) and \( \alpha_c, \alpha_m \) are the coefficients of thermal expansion of cylinder and matrix. The final or effective radius of the cylinder under the constraint of the matrix is \( r_1 \). Equation 7.8 has to be solved with the boundary conditions for the cylinder:

\[
u_r^c(r = R) = r_1 - r_c, \quad u_r^c(r = 0) = 0, \] (7.9)

and for the matrix:

\[
u_r^m(r = R) = r_1 - r_m, \quad u_r^m(r \to \infty) = 0. \] (7.10)

Since the difference \( R - r_1 \) due to contraction is very small, the boundary conditions may be taken into account for simplicity at the boundary \( r = R \). The solution of eq. 7.8 satisfying eqs. 7.9 and 7.10 is

\[
u_r^c(r) = \frac{r_1 - r_c}{R} r, \quad u_r^m(r) = \frac{R(r_1 - r_m)}{r}. \] (7.11)
Using eqs. 7.6 one obtains

\[ \begin{align*}
\epsilon_{\tau \tau} &= \frac{r_1 - r_c}{R}, \\
\epsilon_{\phi \phi} &= \frac{r_1 - r_c}{R} \\
\epsilon_{m\tau} &= -\frac{R(r_1 - r_m)}{r^2}, \\
\epsilon_{m\phi} &= \frac{R(r_1 - r_m)}{r^2}
\end{align*} \]  

(7.12)

The eqs. 7.4 for the two-dimensional case are

\[ \begin{align*}
\sigma_{\tau \tau} &= \frac{E}{1 - \nu^2} (\epsilon_{\tau \tau} + \nu \epsilon_{\phi \phi}) \\
\sigma_{\phi \phi} &= \frac{E}{1 - \nu^2} (\epsilon_{\phi \phi} + \nu \epsilon_{\tau \tau})
\end{align*} \]  

(7.13)

Using eq. 7.13 and imposing \( \sigma_{rr}(R) = \sigma_{r\tau}(R) \) one obtains \( r_1 \). Substituting the latter into eqs. 7.12 and using the definition of \( r_m \) and \( r_c \) yields

\[ \begin{align*}
\epsilon_{\tau \tau} &= \epsilon_{\phi \phi} = \frac{E_m (1 - \nu_c) (\alpha_m - \alpha_c) \Delta T}{E_m (1 - \nu_c) + E_c (1 + \nu_m)} \\
\epsilon_{m\tau} &= \epsilon_{m\phi} = \frac{E_c (1 + \nu_m) (\alpha_m - \alpha_c) \Delta T R^2}{E_m (1 - \nu_c) + E_c (1 + \nu_m) r^2}
\end{align*} \]  

(7.14)

The stress components can be easily found using eqs. 7.13. Figure 7.1 shows the deformation of a lattice calculated with a negative value of \( (\alpha_m - \alpha_c) \Delta T \). The displacements are considered in the plane perpendicular to the cylinder. The pattern far from the cylinder section corresponds approximately to the undistorted grid. A largely exaggerated value of 60\% for \( (\alpha_c - \alpha_m) \Delta T \), with \( E_c/E_m \approx 3 \), was chosen to better show the characteristics of the field.

### 7.3 Axially symmetrical strains with surface relaxation

We consider now a cylinder of radius \( R \) with a finite length \( 2 \ell \ll R \) immersed in a thin plate of infinite radius. The axis of the cylinder is perpendicular to the plate of thickness \( 2 \ell \). In this case, the strains \( \epsilon_{zz} \) and \( \epsilon_{rz} \) (see eqs. 7.6) with both eqs. 7.5 have to be considered, too. The problem may be solved using a so-called Love's function \( \Phi \) [71]. If we define

\[ \begin{align*}
\nu_r &= -\frac{1}{1 - 2\nu} \frac{\partial^2 \Phi}{\partial r \partial z} \\
\nu_z &= \frac{2(1 - \nu)}{1 - 2\nu} \Delta \Phi - \frac{1}{1 - 2\nu} \frac{\partial^2 \Phi}{\partial z^2}
\end{align*} \]  

(7.15)

the equilibrium conditions 7.5 together with eqs. 7.6 and 7.4 are satisfied when the bipotential equation is fulfilled [71]:

\[ \Delta \Delta \Phi = 0 \]  

(7.16)
Figure 7.1:
Lattice distortion due to compressive thermal stress in the plane perpendicular to the cylinder. For simplicity only one quadrant of the cylinder section is shown.

The eq. 7.16 can be solved by means of Hankel transforms [72]. Since

$$\int_0^\infty r\Delta J_0(\xi r)\,dr = \left(\frac{d^2}{dz^2} - \xi^2\right)^2 \int_0^\infty rJ_0(\xi r)\,dr$$

(7.17)

eq \frac{\xi^2}{\xi^2 - \xi^2} \int_0^\infty rJ_0(\xi r)\,dr

(7.18)

eq \frac{\xi^2}{\xi^2 - \xi^2} \int_0^\infty rJ_0(\xi r)\,dr

(7.19)

where $G(\xi, z)$ is the zero-order Hankel transform of the function $\Phi(r, z)$:

$$G(\xi, z) = \int_0^\infty rJ_0(\xi r)\,dr$$

(7.18)

where $G(\xi, z)$ is the zero-order Hankel transform of the function $\Phi(r, z)$:

$$G(\xi, z) = \int_0^\infty rJ_0(\xi r)\,dr$$

(7.19)

The integration of eq. 7.18 leads to the expression

$$G(\xi, z) = (A + Bz)e^{-\xi^2} + (C + Dz)e^{\xi^2}$$

(7.20)

where $A$, $B$, $C$, and $D$ are determined from the boundary conditions. The displacements $u_r$ and $u_z$ may be found using [72]:

$$u_r = \frac{1}{1 - 2\nu} \int_0^\infty \xi^2 \frac{dG}{dz} J_1(\xi r)\,d\xi$$

$$u_z = \int_0^\infty \xi \left(\frac{d^2G}{dz^2} - \frac{2(1 - \nu)}{1 - 2\nu} \xi^2 G\right) J_0(\xi r)\,d\xi$$

(7.21)
In the considered system with $\ell \ll R$, the thermal strains are mainly constituted of plane strains $\varepsilon_{rr}$ and $\varepsilon_{\psi\psi}$, with small additional components $\varepsilon_{zz}$ and $\varepsilon_{rz}$ along the cylinder axis in $z$ direction. Thus, the strain field may be approximated by superposing the solution 7.14 with the strains due only to the thermal contractions of cylinder and matrix along the $z$ axis. The latter strains may be calculated approximately assuming a uniaxial pressure $P$, proportional to the thermal misfit, distributed uniformly over the circular area of the cylinder. If the matrix and the cylinder have the same elastic constants $E$ and $\nu$ the problem is solved using eq. 7.20. The boundary conditions on top and bottom of the cylinder are:

$$\sigma_{zz}(r, z = \pm \ell) = p(r), \quad \sigma_{rz}(r, z = \pm \ell) = 0$$

where:

$$p(r) = \begin{cases} P & \text{if } r < R \\ 0 & \text{otherwise} \end{cases}$$

(7.22)

The Hankel transform of $p(r)$ is

$$\tilde{p}(\xi) = \int_0^R P r J_0(\xi r) dr = \frac{P R J_1(R \xi)}{\xi}$$

(7.23)

Applying the Hankel transform to the conditions 7.22, $A$, $B$, $C$, and $D$ in eq. 7.20 may be determined and $G(\xi, z)$ becomes

$$G(\xi, z) = 2(1+\nu)(1-2\nu) \tilde{p}(\xi) \frac{[\xi^2 \cosh(\xi \ell) + 2\nu \sinh(\xi \ell)] \sinh(\xi z) - \xi z \sinh(\xi \ell) \cosh(\xi z)}{E \xi^3 [2\xi \ell + \sinh(2\xi \ell)]}$$

(7.24)

Inserting eq. 7.24 into the eqs. 7.21 yields the displacements $u_r$ and $u_z$. The latter equations can be evaluated by numerical integration. The pressure $P$ is given by:

$$P = E \varepsilon_{zz} = E(\alpha_c - \alpha_m) \Delta T$$

If the matrix and the cylinder have different elastic moduli the problem cannot be solved this way. Assuming that, in general, $E_c > E_m$, the solution for the matrix is a strain field between the case with equal elastic moduli and that with a very hard cylinder ($E_c \gg E_m$). The latter case can be approximated using eqs. 7.24 and 7.21 with $P$, such that the condition $u_z(r = R, z = \ell) = \int_0^\ell \varepsilon_{zz} dz = \ell (\alpha_c - \alpha_m) \Delta T$, with a constant $\varepsilon_{zz}^c$ within the hard cylinder, is fulfilled. In general for the considered geometry with $\ell \ll R$, we may have differences in $u_z$ of a factor 2 between the two limiting cases. However, in the present composite the difference in the elastic moduli is not that accentuated ($E_c/E_m \approx 3$), thus, the above formulation (eqs. 7.24 and 7.21), using the elastic modulus of the matrix $E_m$, yields a good approximation for the strain field outside the cylinder.

In Fig. 7.2 the distortion of the matrix around a cylinder, calculated with a negative value $(\alpha_m - \alpha_c) \Delta T$, is shown. For the strain evaluation only the thermal contraction
along the z axis was considered, using the eqs. 7.21. A very large \( |(\alpha_c - \alpha_m)\Delta T| \sim 30\% \) was assumed, to accentuate the form of the deformed matrix. The thickness of the plate is 200 nm and the radius of the cylinder 1 \( \mu \text{m} \). The displacements are shown in a positive half-plane containing the cylinder axis. The cylinder is not visible in the figure, its position has to be thought near the left side. In its vicinity the matrix is expanded into z direction following the relative elongation of the cylinder. The expansion decreases approximately as \( 1/r \). The grid far from the cylinder, on the right, corresponds approximately to the undistorted lattice. The radial displacement increases as a function of \( z \), and close to the surface the radius of the cylinder is larger than in the center. In three dimensions the cylinder-matrix interface may be thought of as a kind of hyperboloid parallel to the z direction.

![Figure 7.2: Lattice distortion due to compressive thermal stress in the rz plane.](image)

For simplicity only the positive half-plane is considered. The cylinder is to be thought near the left margin.

### 7.4 Atomistic model

To estimate the surface relaxation of the matrix around the fiber, an atomistic approach was considered, too. The undistorted starting material is described by a periodic lattice. The fiber is defined by a cylindrical region of the lattice where all sites are occupied by species of the type A. The matrix region is defined by species of the type B. An algorithm compares at every site the distance of the neighboring species with a given value and determines in which direction the unit at the considered site has to be moved. The following equation is used to calculate
the new position $\bar{x}'$ of every unit:

$$\bar{x}' = \bar{x} + \frac{1}{N} \sum_{i=1}^{N} (\bar{x}_i - \bar{x}) \frac{|\bar{x} - \bar{x}_i| - r_i}{|\bar{x} - \bar{x}_i|}$$  \hspace{1cm} (7.25)$$

where $N$ is the number of neighbors considered, $\bar{x}$ is the current position, $\bar{x}_i$ are the neighboring positions, and $r_i$ is the distance of the neighbor in the ideal lattice. The latter is obtained under free thermal constraints for the lattices of the single species. The ideal lattices of matrix and cylinder differ according to the thermal misfit. A free boundary is assumed at the surface and far from the cylinder. For the isotropic case, we consider twelve $\langle 110 \rangle$ and six $\langle 100 \rangle$ next-neighbors. After applying recursively eq. 7.25 the units reach a stable position, and the resulting lattice indicates the displacements due to the given thermal stress. In the formalism of eq. 7.25 the elastic constants are not incorporated. Figure 7.3 shows a simulation for $|(\alpha_c - \alpha_m)\Delta T| \sim 10\%$ and equal elastic constants.

![Figure 7.3: Simulation showing the distortion of the lattice, describing the fiber in the matrix under thermal strains.](image)
8 LACBED

8.1 Simulation of LACBED patterns

As explained in chapter 4, in a single LACBED pattern the HOLZ lines in the reciprocal space are related to the image of the sample in real space. Each incident beam direction corresponds to a different column of the sample. The intensity for every incident beam direction is calculated using the lattice parameters of the corresponding sample volume. Beams with different incidence on the specimen are assumed to be incoherent (see [73] for more details): no interference effects between electron waves, generated in the sample by scattering from two different directions of incidence, are considered. The assumption is justified for LACBED, where every beam direction is associated with one place, if no overlapping spots are present. The influence of diffuse intensities due to defects or discontinuities in the crystal can be neglected. The calculation of the LACBED intensities relies on the so-called column approximation [57]. In this approach, the volume of the sample is divided into columns along the beam direction. It is assumed that over the diameter of the column the contribution of each reflection to the image contrast does not change. According to Huygens' principle [75], path differences of the beams less than \( \lambda/2 \) cause no local contrast variations. Thus the radius \( \rho \) of a column depends on specimen thickness \( t \) and wavelength \( \lambda \): \( \rho = \sqrt{t\lambda} \). For the image intensity at one point, only the volume of its surrounding column gives a contribution. The HOLZ lines and the sample image in LACBED patterns taken under optimal conditions, are usually not perfectly sharp, owing to the finite size of the electron source (see Fig. 4.2). The same incident beam direction is associated with a finite area, and not only with a single column on the sample. Furthermore, every point is probed by more than one incident beam direction. Thus, in the simulation of LACBED patterns, a coherent summation of the electron waves over finite areas of the sample and different incident directions should be made. The area is delimited by the used spot size \( S \), while the angular range \( \theta \) to be considered depends on the defocus \( \Delta f \); \( \theta = S/\Delta f \). If a field emission gun (FEG) is used, smallest spot sizes in the nanometer range are available, increasing considerably the sharpness of the pattern. For simplicity we assume in the LACBED simulations that the source is infinitely small.

The intensities in a LACBED pattern are calculated in every sample position, which is associated with a certain incident direction. Given a position \( (x, y) \) on the sample, the incident direction may be defined by a vector \( (x, y, -L) \), where \( L \) is a parameter to be adjusted according to the convergence of the beam. The wave vector \( \vec{k} \) of the incident beam is related to the deviation parameters \( s_g \) of the different reflections \( \vec{g} \).
included in the simulation,
\[ s_g \approx \frac{|\vec{k}|^2 - (\vec{k} + \vec{g})^2}{2|\vec{k}|} \]  \hspace{1cm} (8.1)

In the kinematical approach, the wave amplitude for a reflection \( \vec{g} \) along a column of thickness \( t \), with a displacement \( \vec{u}(z) \) relative to the ideal lattice, is given by:
\[ \psi_g = \frac{i\pi}{\xi_{\vec{g}}} \int_0^t \exp\left\{ -2\pi i[sz + \vec{g} \cdot \vec{u}(z)] \right\} dz \]  \hspace{1cm} (8.2)

The function \( \vec{u}(z) \) may be approximated by \( N \) constant vectors \( \vec{u}_n \) in small thickness intervals \([t_n, t_{n+1}]\), and the total intensity \( I \) is determined with
\[ I = \sum_{\vec{g}} \left( \sum_{n=0}^{N-1} \left( e^{-2\pi i\vec{g} \cdot \vec{u}_n} \int_{t_n}^{t_{n+1}} e^{-2\pi i sz} dz \right) \right)^2 \]  \hspace{1cm} (8.3)

After the incoherent summation for the final LACBED image, all intensities are rescaled and inverted to obtain the defect HOLZ lines in the central disc.

In the dynamical approach, the LACBED simulations are calculated with a modified version of the program used for weak-beam contrast simulations. The formalism shown in chapter 4, in particular eq. 4.16, is used. The function \( \vec{u}(z) \) is also assumed to be constant for each small step in the beam direction, for a better time efficiency of the program. Usually a maximum step size of 10 nm and a minimum step size of 1 nm were taken. The actual selection of the local step size is adjusted according to the local change of \( \vec{u} \) along \( z \).

In eq. 8.2, for the kinematic case, and eq. 4.13, for the dynamical case, a lattice displacement is introduced, according to eq. 4.12, as an additional phase shift \( e^{-2\pi i\vec{g} \cdot \vec{u}} \) in the electron wave. The description of the crystal potential in eq. 4.12 is an approximation for small local distortions. A discrete set of reflections \( \vec{g} \) is sufficient to describe a periodic potential. If distortions are present, the crystal potential has to be formulated as a Fourier integral over a continuous spectrum of scattering vectors in the reciprocal space. A possible way to overcome this problem is to consider a local change of the lattice constants, using a vector \( \vec{g} \) in eq. 8.1 and 4.13, that depends on the position in the sample.

In the kinematic case, for every point of the sample, distorted unit cell parameters may be calculated from the displacements. In the coordinate system of a cubic unit cell, \( \vec{a}_1, \vec{a}_2, \vec{a}_3 \), using the partial derivatives \( \frac{\partial u_i}{\partial x_j} \), with \( i, j = 1, 2, 3 \), one can find the basis vectors of the distorted cell \( \vec{a}'_1, \vec{a}'_2, \vec{a}'_3 \):
\[ \vec{a}'_j = \vec{a}_j + \sum_{i=1}^{3} \frac{\partial u_i}{\partial x_j} \vec{a}_i \hspace{1cm} j = 1, 2, 3 \]  \hspace{1cm} (8.4)
From the distorted unit cell, a new reciprocal lattice and new \( \vec{g} \) vectors can be determined. The deviation parameter \( s \) is found using eq. 8.1 and inserted into 8.2 without the displacement term \( \vec{g} \cdot \vec{u} \). Along the same column the contributions of the individual thickness slices are summed coherently.

To illustrate the difference between the approach with constant and variable \( \vec{g} \) vectors, we consider the strain field caused by a fiber inclined with respect to the sample normal, neglecting surface relaxation. Figure 8.1 shows kinematical LACBED simulations with constant (left) and variable (right) \( \vec{g} \) vector. The diameter of the fiber is 100 nm, the HOLZ reflection is \((791)\), in the \([113]\) zone axis of Si, and the thickness 530 nm. The gray area on the left part of the simulation corresponds to the projection along the zone axis of a fiber. The fiber length is delimited by the top and bottom surface of the sample.

For the factor \( f \) that determines the magnitude of the strain, defined as

\[
f = \frac{E_c(1 + \nu_m)(\alpha_m - \alpha_c)\Delta T}{E_m(1 - \nu_c) + E_c(1 + \nu_m)}
\]

(see eq. 7.14), a value of 0.003 is chosen. The HOLZ line in the simulation on the left in Fig. 8.1 seems to be split in the vicinity of the fiber. The first intensity minimum closer to the fiber becomes lower and lower, while the main one vanishes. The other minima are also affected near the splitting region. In the simulation with variable \( \vec{g} \) vector, there is a similar tendency to splitting, but additional minima and background intensities are present. The subsidiary minima delineating the split, are interrupted in several places. The results of the two simulations are quite different, although the correction with a variable \( \vec{g} \) vector should not influence the main features of the pattern.

In the dynamical simulations, the interaction of every diffracted beam with different sets of \( \vec{g} \) vectors, depending on the position, should be considered, and this would make the computation very complicated. A valid alternative to the Bloch wave formulation would be the so-called multislice method which is well suited for computations involving large numbers of diffracted beams, as is required for simulations of defects or distortions in crystals [74]. An approximation for the Bloch wave approach consists of using different sets of \( \vec{g} \) vectors from column to column, but a constant set within each column. The relative displacements in the column are described by a vector \( \vec{u}' \) referred to the lattice corresponding to the new set of \( \vec{g} \) vectors. We choose a point \( \vec{P} \) in the column and assume a modified unit cell with lattice parameters \( a'_1, a'_2, a'_3 \), and angles \( \alpha'_{12}, \alpha'_{13}, \alpha'_{23} \), where \( \frac{\partial_x u_i}{\partial_x} = \frac{\partial_x u}{\partial_x}(\vec{P}) \), \( \alpha_{ij} - \alpha'_{ij} = \frac{\partial u}{\partial x_j}((\vec{P}) + \frac{\partial u}{\partial x_i}(\vec{P})) \), and \( i, j = 1, 2, 3 \). The vector \( \vec{g} \) is calculated from the latter cell, and it is constant within the whole column. The modified displacements
Figure 8.1:
Kinematical LACBED simulations with a fiber diameter of 100 nm, using a constant \( \bar{g} \) (left), and a variable \( \bar{g} \) (right), with a strain factor \( f = 0.003 \) (see eq. 8.5) and 530 nm thickness.

\( \vec{u}' \) relative to the new cell (see Fig. 8.2) are given by

\[
u'_i(x_i) = u_i(x_i) - \sum_{j=1}^{3} \frac{\partial u_j}{\partial x_j}(P) x'_j, \quad i = 1, 2, 3
\]  

(8.6)

where \( x'_i \) are coordinates with respect to the point \( P \), i.e. \( \vec{x}' = \vec{P} + \vec{x}' \). Since the diffracted intensity depends only on relative changes of the term \( \bar{g} \cdot \vec{u} \), the components \( x'_i \) in eq. 8.6 may be substituted by \( x_i \).

In the dynamical case, as in the kinematic one (see eq. 8.2), the phase of the diffracted electron wave depends on \( s \), \( \bar{g} \) and \( \vec{u} \), combined in \( 2\pi(sz + \bar{g} \cdot \vec{u}) \). Thus, different choices of the reference point \( P \) produce different results. The components of \( \bar{g} \) are inversely proportional to the lattice parameters \( a'_1, a'_2, a'_3 \) whereas the component \( u_i \) depends on the strain. The deviation parameter \( s \) is a function of both the lattice parameters and the wavelength, thus it is not directly correlated with the term \( \bar{g} \cdot \vec{u} \), which does not depend on the electron wavelength. For our simulations, we consider a set of lattice parameters \( a'_{i}(x, y) \) and \( \alpha'_{i}(x, y) \) averaged along the column at position \((x, y)\).

Figure 8.3 shows four LACBED simulations calculated with the dynamical formalism. A fiber diameter of 100 nm, the reflection \((\overline{791})\), and \( f = 0.003 \) are chosen.
Displacements $\vec{u}$ (left) and $\vec{u}'$ (right) relative to a crystal with lattice constants $a$ and $a'$, respectively. At the origin of the diagram, $P$, the displacement is equal, according to eq. 8.6, in both reference systems.

Figure 8.2:

The first two images on the left are simulated keeping a constant $\vec{g}$ vector, with a thickness of 530 nm (Fig. 8.3a) and 800 nm (Fig. 8.3b). The HOLZ line, as in the kinematic case, is split near the fiber. For this case the kinematic model is a good approximation. The thickness characterizes the distance between the subsidiary minima and the sharpness of the split. The two images on the right were simulated assuming $\vec{g}$ averaged along the column (Fig. 8.3c) and taken at the top of the column (Fig. 8.3d). The thickness is 530 nm for both simulations. Figure 8.3c is similar to the simulation with a constant $\vec{g}$ (Fig. 8.3a). The essential difference lies in the curvature of the HOLZ line, that is more accentuated in Fig. 8.3c. The simulation in Fig. 8.3c is more realistic than that of Fig. 8.3d. In Fig. 8.3d, the line and the side minima are strongly modified and the pattern is asymmetric.

Figure 8.4a shows a simulation with the same conditions as in Fig. 8.3c, with a vertical fiber. The fiber causes a local shift of the line, that would not be present in a simulation with a constant $\vec{g}$ vector. The displacement $\vec{u}$ along the zone axis (parallel to the vertical fiber) is constant, and only through a variation of the $\vec{g}$ vector, the influence of the strain field due to the fiber can be included. A simulation assuming constant $\vec{g}$ vectors is therefore not correct. Figure 8.4b illustrates the additional effect caused by the surface relaxation of the sample (see eqs. 7.24 and 7.21). The line splits and the intensity of both side minima decreases in the same way. The curvature of the HOLZ line is due to the variation of $\vec{g}$ as in Fig. 8.4a. This splitting
can be well distinguished from that induced by an inclined fiber, where the curvature of the line and the minima invert.

Figure 8.3:
Dynamical LACBED simulations with a fiber diameter of 100 nm and a strain factor $f = 0.003$ (see eq. 8.5), using a constant $\tilde{g}$ (Fig. 8.3a,b), a $\tilde{g}$ averaged along the column (Fig. 8.3c), and a $\tilde{g}$ taken on the top of the column (Fig. 8.3d). In Fig. 8.3b a thickness of 800 nm, for the others of 530 nm, is assumed.

8.2 Evaluation of local strains from LACBED patterns

LACBED images of large Si precipitates were taken in different regions of the sample to evaluate the local strains close to the fibers. The investigations were performed on a Philips CM 200 FEG equipped with a Gatan imaging filter. The use of an energy filter in elastic (zero loss) mode permits sharp HOLZ lines to be obtained and experimental and simulated intensities to be compared, without perturbations caused by inelastic phenomena. Depending on the convergence of the incident beam, it is possible to probe differently sized areas of the sample. This allows short-range and long-range distortions to be analyzed at different scales. In Fig. 8.5 a LACBED pattern along Si[113] with a precipitate is shown. The precipitate has a size of
Figure 8.4:
Dynamical LACBED simulations using the same conditions of Fig. 8.3c, with a vertical fiber. In the figure on the right the surface relaxation of the sample is taken into account.

Figure 8.5:
LACBED image of Si[113] with small precipitate of about 10 nm size.
about 10 nm and causes only a short-range strain field leading to curved HOLZ lines and Kossel-Möllenstedt fringes in its vicinity. The additional asymmetry of the whole HOLZ pattern is caused by sample bending. A dynamical simulation of the LACBED pattern is shown in Fig. 8.6. The particle is assumed to be cylindrical and immersed in the matrix in vertical position. Surface relaxation and sample bending are not taken into account. Since the composition and the structure of the particle are not known, the factor \( f \) to be used in the simulation can not be calculated. A good agreement with the experimental image is obtained with \( f = 0.002 \) and a sample thickness of 320 nm.

In Fig. 8.7 a LACBED image of Si[113] taken in a region close to a Fiberfrax fiber is shown. In the probed area of the sample (100 nm diameter) there are no large local strain variations, and the image is similar to a CBED pattern. However, some HOLZ lines are split and they cannot be analyzed using the methods explained in chapter 6. The boundary of the fiber is about 200 nm away from the examined region. The reinforcement, with a diameter of about 500 nm, gives rise to a long-range strain field, which does not vary strongly at this distance. We assume for Si, \( \alpha = 3 \times 10^{-6} \text{K}^{-1} \) [78], \( E = 168 \text{ GPa} \) [77], \( \nu = 0.22 \) [77], and for Fiberfrax, \( \alpha = 5.3 \times 10^{-6} \text{K}^{-1} \) (mullite) [79], \( E = 104 \text{ GPa} \) [76], \( \nu \approx 0.2 \), with \( \Delta T = -560 \text{ K} \). The strains calculated
using eqs. 7.14, 7.24, and 7.21 are lower than the actual values, because Si is not simply homogeneously distributed around the fiber. A LACBED simulation of the examined pattern is shown in Fig. 8.8. The fiber is to be thought outside the pattern beyond the lower left corner at a distance of 700 nm, and a thickness of 320 nm is assumed. The HOLZ lines following a radial direction relative to the fiber, are not split, whereas those in azimuthal direction undergo the same effect illustrated in Fig. 8.4b, but on a different scale. The magnitude of the strain causing such a splitting is around $2 \times 10^{-3}$. The level of splitting in the experimental and simulated images are comparable; only the first subsidiary minima are sharp, whereas higher-order minima are not so clearly visible. This indicates that the assumed strain is in a right order of magnitude.

Figure 8.9 shows a LACBED pattern taken along Si[111537]. On the left a portion of a Fiberfrax fiber with a diameter of about 1 μm is visible. The splitting in this case is caused by local distortions close to the interfacial region. These local strains do not originate from the whole fiber. Only small grains near the boundary, such as spinel grains, are responsible for such effects. In Fig. 8.10 a simulation corresponding to the above pattern is shown. A thickness of 240 nm is assumed, and an additional small cylindrical particle (not visible in the picture) overlapped by the large fiber, is also considered in the calculation to simulate the effect of small spinel grains at the surface of the reinforcement. We assume for simplicity that these grains have
LACBED simulation of Si[113] assuming a vertical fiber with 500 nm diameter and with the center 700 nm away from the examined region beyond the lower left corner. The probed area has a diameter of about 100 nm.

LACBED pattern of Si[11 15 37] with part of a Fiberfrax fiber (~ 1 μm diameter) on the left.
LACBED simulation assuming an additional strain field caused by a cylindrical (10 nm radius) spinel grain at the boundary of the fiber. In the figure the small particle is hidden by the fiber.

a circular section, constant in the thickness. The small particle (10 nm radius) is positioned near the interface and is inclined with respect to the sample normal. The strain is calculated using eq. 7.14. Assuming for spinel, $\alpha = 25 \times 10^{-6} K^{-1}$ [80], $E = 385$ GPa, and $\nu = 0.25$ [81], one obtains approximately $f \approx 0.01$, which is larger than for Fiberfrax with $f \approx 0.001$. A splitting similar to that observed in the experimental image is obtained. The form of the splitting is characterized by the inclined particle, similar to the simulations of Fig. 8.3c. The strain due to the whole fiber is superposed to the local strain and induces a curvature in the HOLZ lines in the upper part of the pattern. Here, the influence of the whole fiber is not so strong as in Fig. 8.7 because the ratio between the fiber radius and the thickness is larger and a different long-range displacement field is produced.

Another example of locally concentrated strains is shown in Fig. 8.11. The examined region corresponds to a portion of the lower part of Fig. 8.9. The pattern was taken with a similar beam convergence, but the eccentric height of the specimen was changed to obtain a higher magnification of the sample image. The corresponding simulated pattern in Fig. 8.12 is calculated considering two small cylindrical spinel grains positioned vertically near the boundary of the fiber. The radius of the cylinders is 50 nm and they are separated by a distance of 200 nm. The kind of splitting
Figure 8.11:
LACBED pattern of Si[11 15 37] with a magnified detail of the interfacial region in Fig. 8.9. The probed area has a diameter of about 300 nm.

Figure 8.12:
LACBED simulation assuming additional strain fields caused by two cylindrical (50 nm radius) spinel grains at the fiber boundary.
is similar to the case simulated in Fig. 8.4b, but now the HOLZ lines are not really tangential to the fiber, and thus, not so curved. The strain near the fiber is quite strong, about 0.01, and causes an accentuated splitting. Higher-order minima are strongly affected together with the main minimum.

Considering the examples shown above, we can summarize, that the strains in the region close to the reinforcements are characterized by an overlap of a long-range and a short-range field. Long-range strains are caused by the whole fiber. They are not very strong ($10^{-3}$ or less), and are distributed over a large area, of linear dimensions in the order of magnitude of the fiber size. The short-range strains are produced by the small spinel grains at the fiber boundary and are locally restricted to a region of about 100 nm from the source. The magnitude of the latter strains reaches values up to $10^{-2}$. Since they are very localized, they can be responsible for cracks originating at the interface.
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