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In-situ investigations of structural changes during cyclic loading of aluminium by high resolution reciprocal space mapping

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A major failure reason for structural materials is fatigue-related damage due to repeatedly changing mechanical loads. During cyclic loading dislocations self-organize into characteristic ordered structures, which play a decisive role for the materials lifetime. These heterogeneous dislocation structures are identified by high resolution reciprocal space mapping. The synchrotron technique using high energy x-rays was applied successfully in-situ during cyclic deformation of macroscopic aluminium samples at the Advanced Photon Source to reveal the structural reorganization within single grains embedded in the bulk material during cyclic deformation. As evident from the changes in the radial profiles of four grains, the adaption of the deformation structure to cyclic deformation is completed already after 800 cycles. Individual subgrains have been followed through a 7350 tension-tension cycles while monitoring macroscopic stress and strain during cyclic loading. The elastic back strains of subgrains are Gaussian distributed with larger subgrains showing larger back strains. The detailed characterization of the microstructure during cyclic loading by in-situ monitoring of the internal structure within individual grains facilitates the understanding of materials behaviour during cyclic deformation.

Keywords: cyclic deformation; fatigue; in-situ x-ray diffraction; reciprocal space mapping; aluminium
1. Introduction

The majority of metallic components fail as a consequence of periodically varying stresses causing structural changes in the material, which result in cracks and fracture after a sufficient number of cycles. During mechanical loading of metals, plastic deformation occurs on the microscale by motion of dislocations causing a fraction of dislocations to be stored in the material. Characteristic low-energy dislocation structures develop during cyclic deformation in face-centered cubic metals and consist of dislocation-rich walls and dislocation-free subgrains (Mughrabi et al. 1983). These structures have been extensively studied in copper, while the corresponding microstructural changes in aluminium are less frequently reported. Grosskreutz et al. (1963) and later Madhoun et al. (2003) analysed the reorganization of dislocations in aluminium into 1 µm to 5 µm large cells during cycling deformation by means of transmission electron microscopy. Dislocation walls evolving during cyclic deformation of polycrystals subdivide the original grains into numerous subgrains, i.e. dislocation-free regions of slightly different orientations, as shown for aluminium in figure 1, where the different orientations are revealed by different intensity. The details of the progressing self-organization of dislocations into dislocation walls separating subgrains, however, remain unknown, because it is not possible to study the substructure of grains in the bulk of a relevant sized polycrystal during ongoing deformation by means of electron microscopy. Such information can be provided by sophisticated synchrotron technique while other techniques such as electron microscopy or conventional x-ray diffraction are either restricted to surface near regions, destructive or obtain solely an average over a number of grains with various orientations.

High Resolution Reciprocal Space Mapping (HRRSM) (Jakobsen et al. 2006, 2007) enables to follow the microstructure of individual grains embedded within a polycrystalline bulk sample in-situ during deformation by obtaining three-dimensional reciprocal space maps with high resolution (Δq/q = 10⁻⁴ for the diffraction vector q). Utilizing a custom-made load frame, the evolution of the subgrains and the associated internal stresses in individual grains of commercially pure, polycrystalline aluminium can be monitored in-situ during cyclic deformation. In this manner, the evolution of deformation structure can be related in an unprecedented way to the mechanical loading experienced by the sample, for instance, during unloading and reversed loading (Diederichs et al. 2017a) or after an increasing number of load cycles (Diederichs et al. 2017b). In the present study, four grains of similar orientation (having a crystallographic (100) direction along the loading axis) are followed through an increasing number of tension-tension load cycles. In each of the grains, at least 80 subgrains
are identified from their corresponding high-intensity peaks and their evolution during cyclic deformation is analysed.

Fig. 1. Deformation structure within an individual grain in polycrystalline aluminium (AA1050) after cyclic deformation in tension-tension for 40500 cycles with a nominal strain amplitude $\dot{\varepsilon}_{\text{nom}}$ of $6.7 \cdot 10^{-4}$. The electron channelling contrast image reveals subgrains of about 2 $\mu$m size by their orientation difference originating from excess dislocation in the dislocation walls.

2. Experimental investigation

2.1. Material

Tensile test specimens were manufactured from an AA1050 sheet homogeneously cold-rolled to 90% thickness reduction to a final thickness of 1 mm. Dog bone-shaped specimens with a gauge section of 15 mm in length and 5 mm in width were designed to fit to a custom-made screw-driven load frame. Sample cutting was done by spark cutting and tensile specimens were then annealed at 600 °C for 2 h to ensure complete and homogeneous recrystallization. The microstructure after annealing was investigated metallographically. Using both, light optical microscopy and scanning electron microscopy, grain sizes were estimated to be between 30 $\mu$m and 100 $\mu$m, and the microstructure was found to be homogeneous throughout the entire cross section of the gauge showing a pronounced cube texture. Tensile testing of such recrystallized specimens with a nominal strain rate of $10^{-3}$ s$^{-1}$ revealed their yield strength being 16 MPa.
2.2. Pre-deformation

Prior to the in-situ investigation by HRRSM, cyclic pre-deformation was carried out in order to introduce a microstructure conform to cyclic deformation in the specimen using an MTS Acumen 3 kN Electrodynamic Test System equipped with Station Manager MTS FlexTest 40 and pneumatic grips. The investigated sample was initially deformed by 1% in tension with a grip speed of 0.015 mm/s and then cycled at a rate of 0.5 Hz under displacement control with a displacement amplitude of 10 µm corresponding to a nominal engineering strain amplitude \( \hat{\varepsilon}_{\text{nom}} \) of 6.7 \( \times \) 10^{-4}. 18000 tension-tension cycles were performed by repeatedly decreasing the displacement by 20 µm from the maximal displacement of 150 µm achieved after tensile deformation to 1% and increasing it again to 150 µm. During the cyclic deformation, a stress ratio \( R = \sigma_{\text{min}}/\sigma_{\text{max}} \) between the stresses at minimum and maximum load of 0.65 was observed.

2.3. Experimental set-up at synchrotron facility

For the synchrotron investigations, the sample was equipped with a pre-wired strain gauge Omega KFG-3 350 Ω at the center of the gauge section and aligned with the tension axis to monitor the axial strain in-situ. The sample is mounted in a custom-made screw-driven load frame equipped with a 5 kN load cell as presented in figure 2a. Using flat grips, this load frame allows for mechanical loading in tension and compression while monitoring the local microstructure within the tensile specimen in-situ using synchrotron radiation. The load frame was placed with the load axis horizontally on a xy-translation stage allowing to move the sample and with this the selected grain of interest – which becomes displaced with respect to the load frame during mechanical loading – to the center of the beam after each loading step. The xy-translation stage is mounted on top of a rotation stage allowing rotation of the entire load frame around the vertical z-axis to obtain reciprocal space maps by rocking in small intervals around this axis. An additional z-translation stage allows adjustment for possible changes in the height of the selected grain of interest due to mechanical loading.

High Resolution Reciprocal Space Mapping was carried out at beam line 1-ID-E at the Advanced Photon Source at Argonne National Laboratory with a monochromatic beam of 52 keV while loading the pre-fatigued sample with position control. A sketch of the experimental set-up is shown in figure 2b.
First, suitable grains are identified with the help of a large area detector, an amorphous silicon flat panel from General Electrics, (detector 1) placed 86 cm behind the sample on a horizontal translation to cover the first 6 diffraction rings of aluminium. Individual grains are selected by finding isolated 400 diffraction peaks from a set of diffraction patterns (acquired at different \( \omega \) angles but at the same sample position to determine grain size and neighbourhood) with corresponding diffraction vector close to the tensile axis and not overlapping with peaks of other grains. After this, the near detector 1 is moved out of the beam and the diffraction peaks are investigated with higher angular resolution by a Mar165 CCD (detector 2) placed 4.65 m behind the sample on the location of 400 diffraction peaks with diffraction vector close to the tensile axis (i.e. in the horizontal diffraction plane at a diffraction angle \( 2\theta_{400} \) of 13.53° for aluminium). An entire sequence of two-dimensional images for each selected diffraction peak is acquired with the far detector 2, while rocking the sample around the vertical \( z \) axis perpendicular to the scattering plane in small intervals \( \Delta \omega \) of 0.015° of the rocking angle \( \omega \). In this way three-dimensional distributions of the diffracted intensity (consisting of the two directions on the detector and the additional rocking) are obtained by stacking the images recorded for several adjacent \( \omega \) intervals. From these three-dimensional reciprocal space maps, structural features such as dislocation walls and subgrains within individual grains can be identified due to the intensity differences caused by slightly different and unique orientation of the subgrains in the
deformation structure as the crystalline lattice becomes locally distorted by dislocation structures. Sharp peaks of high intensity in the high resolution reciprocal space map correspond to individual subgrains, whereas a smooth cloud of lower intensity originates from the dislocations walls (Jakobsen et al. 2007). From the characteristic intensity distribution of ordered dislocation structures, both the distribution of elastic strains within single grains as well as from the embedded individual subgrains can be resolved (Jakobsen et al. 2007, Pantleon et al. 2010). Four individual grains of similar orientation with their crystallographic [001] direction along the loading axis have been selected in the pre-deformed specimen. High resolution reciprocal space maps for each of them have been obtained for 11 different loading steps. The details of the loading steps are summarized in table 1.

2.4. In-situ deformation

The sample was first loaded elastically in uni-axial tension to the highest stress experienced during pre-deformation (load step L0) so that the stress corresponded to the one achieved after 1% pre-deformation in tension. Tension-tension cycling was then performed such that the displacements during cycling were lowered and never exceeded the displacement after the initial uni-axial loading step L0. The sample was cycled for 7155 cycles in total (load steps C1-C3) in position control of the movable cross head. A constant displacement amplitude for the cross head was chosen leading to an effective macroscopic strain amplitude \( \varepsilon_{\text{macr}} \) of 0.8·10^{-4} as measured by the strain gauge (rather than the nominal strain amplitude determined from the cross head displacement). This caused larger stress amplitudes and, consequently, a lower stress ratio \( R \) of 0.29 than during the pre-deformation. As neither the pre-deformation, nor these first cycles caused a significant broadening of the diffraction peaks to the desired extent, the sample was further loaded in uni-directional tension by an additional macroscopic strain \( \Delta \varepsilon_{\text{macr}} \) of 0.1% (L1) and 0.3% (L2), i.e. to a total tensile strain (macroscopic strain plus nominal pre-strain of 1%) of 1.1% and 1.3% respectively, the latter being the highest strain the sample experienced during the entire experiment. The displacement achieved at the end of L2, i.e. after 1.3% strain, served again as maximal displacement of the cross head during the following cycling sequence. A second tension-tension cycling (load steps C4-C8) was then performed under position control of the cross head, by decreasing the displacement by a larger amount corresponding to a macroscopic strain amplitude \( \varepsilon_{\text{macr}} \) of 2.3·10^{-4} and a stress ratio \( R \) of 0.1. This second cycling by in total 7350 cycles (C8) after loading to a total tensile strain of 1.3% (L2) is discussed in detail. During the tension-tension cycling, load cell and strain gauge were monitored with a frequency of 1 Hz. The resulting macroscopic stress strain curves for the last load cycle of each of
the load steps C4-C8 are presented in figure 3. They clearly reveal a mechanical hysteresis evolving only slight changes with increasing number of cycles.

![Macroscopic hysteresis curves obtained during in-situ cyclic loading. Full circles connected by solid lines indicate the stresses and strains recorded during the last cycle of each of the performed cycling loadings (C4 to C8). Due to the read-out frequency of 1 Hz, absolute maxima and minima within the cycles are not always captured; therefore, these are traced from the entire data set and marked with squares in the corresponding color.](image.png)

After each loading step (L0, C1-C3, L1, L2, C4-C8, cf. table 1), high resolution reciprocal space maps for each of the four grains were collected. Each acquisition takes about 15 minutes, depending on the number of necessary Δω intervals to acquire the entire orientation spread developed in the grain. Each rocking interval requires in total 17 s including time for motor movements and an exposure time of 5 s. Usually, between 40 and 60 different rocking intervals were acquired for each reciprocal space map. The time for centering of each grain and acquisition of HRRSM for all four grains after each load step was about two hours. Unfortunately, the intensity of the incoming beam dropped significantly after loading step C8. While analysis of the mechanical data and integrated profiles is reliable, only limited information on the subgrains is available from this acquisition.
Table 1. Designation of load steps for acquisition.

<table>
<thead>
<tr>
<th>Load step</th>
<th>Acquisition after</th>
</tr>
</thead>
<tbody>
<tr>
<td>L0</td>
<td>Pre-deformation to $\varepsilon = 1%$, 18000 cycles, $\dot{\varepsilon} = 6.7 \cdot 10^{-4}$ + Mounting and tensile loading</td>
</tr>
<tr>
<td>C1 – C3</td>
<td>7155 cycles, $\dot{\varepsilon}_{\text{nom}} = 0.8 \cdot 10^{-4}$</td>
</tr>
<tr>
<td>L1</td>
<td>Loading to $\Delta \varepsilon_{\text{macr}} = 0.1%$, i.e. 1.1% total tensile strain</td>
</tr>
<tr>
<td>L2</td>
<td>Loading to $\Delta \varepsilon_{\text{macr}} = 0.3%$, i.e. 1.3% total tensile strain</td>
</tr>
<tr>
<td>C4</td>
<td>800 cycles, $\dot{\varepsilon}_{\text{macr}} = 2.3 \cdot 10^{-4}$</td>
</tr>
<tr>
<td>C5</td>
<td>2650 cycles, $\dot{\varepsilon}_{\text{macr}} = 2.3 \cdot 10^{-4}$</td>
</tr>
<tr>
<td>C6</td>
<td>550 cycles, $\dot{\varepsilon}_{\text{macr}} = 2.3 \cdot 10^{-4}$</td>
</tr>
<tr>
<td>C7</td>
<td>1150 cycles, $\dot{\varepsilon}_{\text{macr}} = 2.3 \cdot 10^{-4}$</td>
</tr>
<tr>
<td>C8</td>
<td>2200 cycles, $\dot{\varepsilon}_{\text{macr}} = 2.3 \cdot 10^{-4}$</td>
</tr>
</tbody>
</table>

3. Experimental results

High resolution reciprocal space maps of all four grains were always acquired in the same sequence starting from grain 1 to grain 4 after cycling. This acquisition sequence differs only after tensile loading (L2), when the sequence 1,3,2,4 was followed by a repetition of grain 1 with a larger range of the rocking angle.

3.1. Macroscopic stress-strain behaviour

During acquisition of each high resolution reciprocal space map, the applied load and the resulting strains were recorded by the load cell and the strain gauge, respectively. The average macroscopic stress and strain during each acquisition are displayed in Figure 4. During pausing of the motors for acquisition of the HRRSM after each loading step, the position of
the cross heads is fixed, while stresses and strains do not remain constant. This is in particular prominent for the acquisition after tensile deformation to a total strain of 1.3% (L2), where the stress drops from 39.6 MPa to 38.6 MPa between the two repeated measurements of grain 1. Simultaneously, the macroscopic strain decreases, indicating a relaxation in the loaded parts of the load frame. After the performed tension-tension cycles a similar behaviour is observed: During the acquisition period for the four grains, a decrease of both the macroscopic axial stress and strain was detected after each load step. These differences of stresses and strains between the first and the last grain measured at each load step decrease with the increased number of cycles. Additionally, an overall decrease in macroscopic stress and strain is observed from after the first tensile loading (L2) to the last cycling (C8) (from 39.6 MPa to 37.2 MPa and from a strain of about 2.94·10^{-3} to 2.90·10^{-3}). Notably, the behaviour of the material after the first cycling (C4) following immediately after the loading step seems to be different from that of the following cycling steps (C5-C8). While the stress decreases continuously, the strain increases during the first cycling (C4) in comparison to the strains after loading (L2), e.g. for the time of acquisition of grain 2 from 2.93·10^{-3} to 2.94·10^{-3}.

**Fig. 4.** Average macroscopic axial stress and strain during the different acquisition steps after loading and cycling for grains 1 to 4. An additional acquisition step has been performed for grain 1, immediately after loading and again after acquiring data sets for grains 2 to 4. The acquisition sequence is indicated by arrows.
3.2. High resolution reciprocal space mapping

The intensity distribution of the 400 diffraction peaks from the four selected grains acquired using the far detector 2 after each load step can be analysed and presented as azimuthal maps or radial profiles, which represent the distributions of lattice plane inclinations and normal strains, respectively. The intensity distribution of each individual diffraction peak is not completely smooth due to local distortions of the crystalline lattice within an individual grain caused by the introduced dislocation structures. These dislocation structures consist of dislocation-free subgrains separated by dislocation walls, where the subgrains cause sharp high-intensity peaks on top a cloud of smooth intensity.

Figure 4 shows the azimuthal projections of the 400 diffraction peak of grain 1 before and after loading to 1.3% tensile strain and after each cycling step (cf. Diederichs et al. 2017b for similar analysis of grain 3). A clear increase in the width of the diffraction peak becomes visible after loading from 1.1% to 1.3% total tensile strain, i.e. from L1 to L2, while the diffraction peak does not undergo such significant changes during the subsequent cycling (C4-C8).

Fig. 5. Azimuthal maps of grain 1 for the load steps L1, L2 and C4 to C8. A clear broadening of the reflection is visible from L1 to L2. The projections do not differ significantly from L2 to C7; after C8 the beam intensity dropped significantly causing a lower intensity and a different signal to noise ratio.
comparison purposes, measurements were normalized with the intensity of the incoming beam and scaled in the same manner (L1-C7);

Fig. 6. Example for partitioning of the intensity distribution: (a) azimuthal map for load step L2 and the two corresponding extracted components, (b) the peak component comprising the high-intensity peaks from the subgrains and (c) the smooth cloud component originating from the dislocation walls.

As discussed in section 2.3, the heterogeneous intensity distribution in the HRSM consists of high-intensity peaks and a spread out cloud of lower intensity. For further analysis, each azimuthal map is partitioned as illustrated in figure 6 into a smooth cloud component and the peak component containing all high-intensity peaks using mathematical approaches (Wejdemann et al. 2011). The cloud of lower intensity represents the dislocation-rich wall component, whereas the peak component constitutes the subgrain component and represents the sum of the diffracted intensities from all subgrains present in the grain. Each high-intensity peak corresponding to a local maximum in the peak component, e.g. in Figure 7, represents a nearly dislocation-free subgrain and can be analysed individually. In this way, individual subgrains can be identified by their corresponding high-intensity peaks and traced from acquisition to acquisition as for example the high-intensity peaks marked in Figure 7 for L2, C4 and C7. In some cases, it cannot completely ruled out that the diffracted intensity may origin from more than one subgrain, in particular, for peaks of lower intensity, (Wejdemann et al. 2011); they will therefore be neglected in the further analysis.

Fig. 7. Azimuthal map of the peak component for the selected load steps L2, C4 (after 800 cycles) and C7 (after 5150 cycles) for grain 1. Four peaks from
four individual subgrains 1-4 found in all load steps (L2-C8) are marked as examples.

Fig. 8. Microstructural development during cycling from L2 to C7: (a) the total integrated (not normalized) intensity of grain 1 in comparison with the extracted intensity of the subgrain component and (b) the subgrain volume fraction for all four grains.

Figure 8a shows the development of the overall measured intensity of grain 1 during the second cycling in comparison to the separated subgrain component after partitioning. The total integrated intensity shows only small variations due to fluctuations in the incoming beam intensity. The intensity of the subgrain component (integrated intensity of the peak component) is about 40% of the total intensity. The volume fraction of subgrains is calculated by dividing the integrated intensity of the peak component by the total integrated intensity. As obvious from figure 8b, all four grains show a constant subgrain volume fraction for all acquisitions taken along the second cycling. The volume fraction of grain 3 is with 70% significantly higher than the volume fraction of 40% as measured for all other grains, which is related to a more narrow appearance of the grain in the reciprocal space map with a lower cloud component allowing an easier separation of the subgrain component and does not indicate a major difference in the deformed structure. The acquisition after load step C8 is not included here due to the significant drop in the beam intensity.

3.3. Radial profiles of the investigated grains

The corresponding radial profiles of grain 1 for L2-C8 are shown in figure 9a. The radial profiles, which are projections of the reciprocal space maps on the direction of the diffraction vector, can be characterized using two variables: By the diffraction angle \(2\theta\) (from Bragg’s equation \(2d_{400}\sin \theta = \lambda\) with the wave length \(\lambda\) of the used X-rays, and the lattice plane
spacing \( d_{400} \) between crystallographic \{400\} planes) or as demonstrated in the following by the modulus of the diffraction vector

\[
q = \frac{4\pi}{\lambda} \sin \theta.
\]

The obtained radial profiles are fitted with a split pseudo-Voigt function and analyzed further regarding their mean position, profile width and asymmetry. Those characteristic parameters provide information on the lattice strain along the diffraction vector. The peak position can be quantified by the average position \( q_{\text{mean}} = 2\pi/d_{400} \) of the profile. \( q_{\text{mean}} \) is inversely proportional to the lattice plane spacing, meaning that a larger lattice spacing results in a smaller \( q_{\text{mean}} \). While the profiles shift to lower \( q_{\text{mean}}^{\text{grain}} \) during the tensile loading of the sample (cf. fig. 9b), the profiles shift to slightly higher \( q_{\text{mean}}^{\text{grain}} \) during the cycling sequence (from black over green to red in fig. 9a) in full accordance with the applied stresses. The shift in peak position during cycling \( (\Delta q_{\text{mean}}^{\text{grain}} = 1.9 \times 10^{-4} \text{ Å}^{-1}) \) is quite small in comparison to the shift during tensile loading \( (\Delta q_{\text{mean}}^{\text{grain}} = 1.2 \times 10^{-3} \text{ Å}^{-1}) \) as visible in figure 9b. The shift of the peak position during the second cycling is exemplarily shown in figure 9c for grain 1; \( \Delta q_{\text{mean}}^{\text{grain}} \) shifts to smaller values after the first cycling after loading and increases during cycling following closely the macroscopic stress. Note that the difference in the mean peak positions between the four grains seen in figure 9c for all load steps is despite their similar orientation caused by the different neighbouring grains in the polycrystalline specimen.
The integral width $\beta^\theta$ of a radial profile is defined as ratio between the total integrated intensity, i.e. the area below the peak in the $q$-range with significant intensity (here 1/50 of maximum intensity), and the maximum intensity. Figure 10a presents the integral width $\beta^\theta$ of the radial profiles for each of the four grains in dependence on the macroscopic strain. A clear increase in the integral width is observed after each tensile loading step (L1, L2), while the detailed display of the second cycling in figure 10b shows a slight decrease in width after the first cycling C4 after tensile loading L2. During further cycling (C4-C8), the integral width does not change significantly any longer, but stays almost constant. Notably, the integral width differs significantly between the four grains.

An asymmetry in the radial profiles indicates the presence of an ordered structure as rationalized by the composite model (Ungar et al. 1984, Mughrabi et al. 1986, Pantleon et al. 2010): asymmetric radial profiles are considered as the sum of symmetric profiles from individual soft subgrains and hard dislocation walls, which develop different elastic strains during deformation and hence diffract at different radial profile positions. Their superposition results in an asymmetric radial profile for the entire grain, where the asymmetry is directly related to the difference in elastic strains between the subgrains and the walls along a direction parallel to the diffraction vector. The absolute asymmetry $\kappa$ of the radial profiles is defined as the difference between the width on the low and the high $q$-side of the maximum intensity, where the width on each side of the peak maximum $q_{\text{max}}^{\text{grain}}$ is determined by the position at which half of the maximum intensity is attained. The values obtained by fitting split pseudo-Voigt functions to the experimental data are shown in Fig. 11a for all four investigated
grains. For grains 2-4, the absolute asymmetry $\kappa$ increases significantly during the first tensile loading from $2 \cdot 10^{-4}$ Å$^{-1}$ to $4.8 \cdot 10^{-4}$ Å$^{-1}$ and for grains 3 and 4 also during the second tensile loading to a maximum of $6 \cdot 10^{-4}$ Å$^{-1}$, while it remains almost constant for grains 1 and 2 during the second cycling. Grain 1 starts with a much higher asymmetry of $5.1 \cdot 10^{-4}$ Å$^{-1}$ after pre-deformation and mounting and shows a decrease during the first loading. However, all grains develop a similar absolute asymmetry after L2. Small variations are measured during the cycling itself as shown in detail for load steps L2-C8 in Figure 11b.

The absolute asymmetry is positive for all measurements indicating that the subgrains have larger diffraction vectors and smaller lattice spacings than the dislocations walls. This means that compared to the average of each grain, the dislocation walls experience tensile strains and the subgrains experience compressive strains — in full accordance with the predictions of the composite model for observations along the tensile axis (i.e. in an axial case under tension). In the following, the subgrain component will be analysed in more detail.

---

**Fig. 10.** Integral width $\beta^2$ of the radial profiles for grain 1 to 4 as function of the macroscopic strain: (a) for all load steps, and (b) for the second cycling L2-C8 after loading to 1.3% total tensile strain.
3.4. Individual Subgrains

In order to perform a detailed analysis of the internal structure of the investigated grains, subgrains are resolved from the high resolution reciprocal space maps of each grain by their individual high-intensity peaks. Using an automatic software algorithm (Wejdemann et al. 2010, 2013) the 100 most intense high-intensity peaks are identified for each reciprocal space map; each of these peaks presumed to originate from to a single subgrain. In the less deformed specimen (until the L1), less than these 100 subgrains were found for all grains. After a tensile loading to 1.3% total tensile strain (L2 and the subsequent cyclic loadings C4-C8) at least 100 subgrains were identified, where the 80 most intense and thus largest subgrains will be analysed in the following.

In order to reveal internal stresses and strains, the average peak positions of the subgrain profiles are compared to the average position of the entire grain they belong to. Based on the measured peak shift $\Delta q$ between the subgrain profile and the grain profile

$$
\Delta q = q_{\text{subgrain}}^{\text{mean}} - q_{\text{grain}}^{\text{mean}}
$$

the internal elastic strain $\varepsilon$ of an individual subgrain with respect to the entire grain can be directly calculated

$$
\varepsilon = \frac{q_{\text{subgrain}}^{\text{mean}} - q_{\text{grain}}^{\text{mean}}}{q_{\text{grain}}^{\text{mean}}} = \frac{q_{\text{grain}}^{\text{mean}} - q_{\text{subgrain}}^{\text{mean}}}{q_{\text{grain}}^{\text{mean}}}. \quad (1)
$$

A positive peak shift $\Delta q$ of a specific subgrain profile is for the presented experimental set-up associated with elastic
compressive back strains of the corresponding subgrain.

The relative peak positions of the 80 largest subgrains of grain 1 are shown exemplarily in Figure 10 for L2 and C7 (after 5150 cycles). The mean positions of the individual subgrain profiles are not spread around the mean position of the grain profile and only a few subgrains have diffraction vectors lower than that of the grain average. In general, the radial positions of the subgrains are found at larger diffraction vectors than the average of the grain indicating elastic back strains. Instead of being distributed around the average peak position \( q_{\text{mean}}^{\text{grain}} \) of the entire grain, they are gathering around the position of the peak maximum \( q_{\text{max}}^{\text{grain}} \) of the profile of the entire grain as indicated by the dashed lines representing the peak shift between the position of the maximum intensity and the average position in figure 12. In the normal probability plot shown in figure 12, the data points of each load step follow almost a straight line revealing that the peak shifts \( \Delta q \) of the individual subgrains follow a Gauss distribution (similar to Wejdemann et al., 2013 for a tensile deformed sample). Comparing the two load steps L2 and C7, a shift to slightly higher peak shifts is visible after cycling accompanied by an increase in the slope reflecting a decrease in the spread of the subgrain profile positions. The mean value of the distribution increases from \( 3.6 \cdot 10^{-4} \, \text{Å}^{-1} \) for L2 to \( 4.1 \cdot 10^{-4} \, \text{Å}^{-1} \) for C7, while the standard deviation of the distribution decreases slightly from \( 3.6 \cdot 10^{-4} \, \text{Å}^{-1} \) to \( 3.2 \cdot 10^{-4} \, \text{Å}^{-1} \). For the majority of the subgrains, an increase in the peak shift \( \Delta q \) is expected from Figure 12.

**Fig. 12.** Normal probability plot of the peak shift \( \Delta q \) between the position of subgrain profiles and the mean position of the entire grain for the 80 most intense high-intensity peaks of grain 1 for L2 and C7. Best linear fits to the data are shown as lines indicating that the peak positions follow a Gaussian
distribution around the position $q_{\text{grain}}^{\text{max}}$ of the peak maximum with a narrower spread after cycling. The peak shift $\Delta q_{\text{grain}}^{\text{max}} = q_{\text{max}}^{\text{grain}} - q_{\text{mean}}^{\text{grain}}$ of the maximum intensity of the corresponding radial grain profile is marked as a dashed line.

Four high-intensity peaks (as marked in figure 7) were identified in all azimuthal projections of grain 1 and followed individually for all load steps from L2 to C8. These four high-intensity peaks were chosen among the ten highest intensities and are, hence, four of the largest subgrains identified in grain 1. In figure 13, the radial profiles of the four selected subgrains are shown together with the corresponding radial profiles of grain 1 (thin lines) for load steps L2-C8. According to figure 9a the grain profile shifts to higher diffraction vectors during cycling. For all four subgrains, the profiles shift to higher $q$-values with cycling as well and follow the grain behaviour (from black to red).

The integrated intensity calculated from the radial subgrain profiles (of the normalized data) for each load step is shown for the four selected subgrains in figure 14a, underlining that it was possible for these four subgrains to trace their individual position and intensity after each load step. The intensity varies for most of the acquisitions between $2.5 \cdot 10^4$ counts and $5.5 \cdot 10^4$ counts and is in average the largest for subgrain 1 and the smallest for subgrain 4 indicating that subgrain 4 is smaller (i.e. has a smaller volume) than subgrain 1. From their detected intensity, all four subgrains are expected to be of comparable size and their size remains nearly unchanged during all load steps (confirming that it is indeed the same subgrain).

Figure 14b shows the development of the peak shift $\Delta q$ between the mean position of the subgrain and the entire grain profile. After tensile loading (L2), subgrains 1 and 2 differ significantly in their peak shift ($\Delta q = 3.4 \cdot 10^{-4}$ Å$^{-1}$ and $\Delta q = 6.9 \cdot 10^{-4}$ Å$^{-1}$) in 3.5·$10^{-4}$ Å$^{-1}$, while the profiles of subgrain 3 and 4 have comparable peak shifts ($\Delta q = 4.73 \cdot 10^{-4}$ Å$^{-1}$ and $\Delta q = 4.76 \cdot 10^{-4}$ Å$^{-1}$) in between these values. The subgrain profiles for subgrain 1, 3 and 4 develop larger shifts $\Delta q$ leading in general to higher peak shifts after 5150 cycles (C7) than after L2 with largest gains for subgrains 1 ($2.1 \cdot 10^{-4}$ Å$^{-1}$) and 4 ($1.8 \cdot 10^{-4}$ Å$^{-1}$). Subgrain 2 which had the largest peak shift after L2 nearly kept this value and decreased the peak shift only slightly.

In summary, the peak shifts of all four subgrains converge to more similar values after C7 in comparison to their starting values after L2, which is also indicated by the narrower spread of the peak shift distribution of all 80 subgrains of C7 in
comparison to L2 discussed in figure 10. The dashed black line represents the mean peak shift of all 80 subgrains in the analysed grain 1, which is in general smaller than the individually presented peak shifts for the four selected and large subgrains. Figure 14c shows similar trends for the internal elastic strain calculated after equation (3). The experienced elastic strain is always negative, i.e. a back strain. Initially (after L2) the values are quite different from subgrain to subgrain ($\Delta \varepsilon = 6 \cdot 10^{-5}$), but more similar values ($\Delta \varepsilon = 2 \cdot 10^{-5}$) are adapted after C7 indicating a levelling of the elastic strains in the cyclic deformation structure.

The elastic back strains of the four selected subgrains, which were among the largest, are in general higher than the mean elastic strain of all 80 subgrains. This gives an indication that the larger subgrains experience a higher elastic back strain than the majority of (smaller) subgrains in the grain. To provide more evidence for this size effect the variation of the elastic strains experienced by the individual subgrains as determined from the peak shift of their corresponding high-intensity peak is shown in figure 15a for the 80 largest subgrains of all load steps L2 to C7. The 80 peaks with highest intensity from each load step were grouped in three groups of intense (corresponding to the 10 largest subgrains, green), medium intense (corresponding to the 20 medium sized subgrains having smaller sizes than the 10 largest ones, red) and low intense (the 50 smallest of the 80 considered subgrains, blue) peaks. Only few high-intensity peaks reach intensities up to $9 \cdot 10^5$ counts and can be related to exceptionally big subgrains. The majority of the peaks has intensities between $1 \cdot 10^5$ counts and $3 \cdot 10^5$ counts meaning that the majority of subgrains are of comparable size. Most of the subgrains experience negative elastic strains related to their positive peak shift $\Delta \varepsilon$ as already seen from figure 12. The average elastic strain experienced by the subgrains of each of these three groups was calculated and marked with a coloured dashed line. These lines reveal the presence of a clear size effect, the largest subgrains (green) experience in average an elastic back strain, which is two times higher than that of the smaller subgrains (blue). For a more detailed assessment of this size effect, the data from the high-intensity peaks for the identified subgrains in grain 1 after L2 and C7 are ordered with respect to their intensity in Figures 15b and 15c. Figure 13b shows the decreasing intensity of the peaks proportional to the volume of subgrains. In addition, the peak shifts $\Delta \varepsilon$ of the subgrains from L2 and C7 are shown in the same order in Figure 13c, revealing a tendency for the less intense peaks originating from smaller subgrains to have lower peak shift $\Delta \varepsilon$ and hence a lower elastic back strain.
Fig. 13. Radial profiles of grain 1 and the four selected subgrains 1-4 marked in Fig. 7 for load steps L2 to C8. Both grain and subgrain profiles shift to higher diffraction vectors during cycling. The maximum intensity is normalized to 1 for the grain profile and to 0.5 for each subgrain profile.
Fig. 14. (a) Integrated intensity of the four selected subgrains 1-4 for the load steps L2 to C7. Subgrain 1 corresponds to the most intense peak and thus the largest subgrain. (b) Peak shift \( \Delta q \) for the four selected subgrains and (c) development of the internal elastic strain during acquisition for load steps L2 and C7. The mean peak shift and elastic strain experienced by all 80 subgrains of grain 1 are marked with a dotted black line.

Fig. 15. Analysis of the 80 largest subgrains in grain 1 for load steps L2-C7: (a) Peak intensities and corresponding elastic strains for the 80 largest subgrains of each load step divided in three groups according to their order with respect to their peak intensity (1:10 in green, 11:30 in red and 31:80 in...
blue). The average elastic strain has been marked for each group with a dashed line of same colour, indicating that larger subgrains experience in average a higher elastic back strain than smaller subgrains. (b) Peak intensities of the subgrains in order of decreasing intensity for load steps L2 and C7. (c) Peak shift $\Delta q$ of the subgrains in order of decreasing intensity for L2 and C7 revealing a slight decrease in $\Delta q$ for decreasing peak intensity of the subgrain profiles.

4. Discussion

4.1. Macroscopic mechanical behaviour

High resolution reciprocal space mapping was performed in-situ after each loading or cycling step (cf. fig. 4) at the same cross head position during all acquisitions while macroscopic stress and strain were monitored. A continuous decrease in the applied stress was revealed during cycling after the final tensile loading step. As the macroscopic strain measured by the strain gauges decreases in a similar manner during the individual acquisition sequence (where the cross head position is kept constant) and during subsequent cycling steps (where the cross head is moved in position control to a smaller displacement and back again), this cannot be attributed to stress relaxation by forward plastic deformation in the specimen, rather stress relaxation occurs in the entire load frame. Even so, the effect of stress relaxation becomes less and less pronounced after more and more cycles (from 1.35 MPa after the first cycling after loading to 0.15 MPa after the last cycling) as well as the associated strain decrease (from $1.95 \times 10^{-5}$ to $1.4 \times 10^{-6}$) indicating a lower sensitivity of the setup towards further stress relaxation. The reason for the increase in macroscopic strain during the first cycling step (i.e. after load step C4) remains unclear, but might have been caused by performing a few cycles interruptedly using small subsequent displacements steps before starting automatic cycling with constant displacement amplitude.

4.2. Evolution of the deformation microstructure during second cycling

After uni-directional tensile loading to a total strain of 1.3% (L2), only minor changes in the azimuthal maps of the diffraction peaks were observed (cf. fig. 5) during the subsequent cyclic deformation in tension-tension sequences, which is in full accordance with expectations from repeated cyclic loading (Essmann et al. 1979, Guichon et al. 1984). The diffracted intensity related to the high-intensity peaks and the volume fraction of the subgrain component in each grain (e.g. about 40% for grain 1) stay almost constant during the second cycling (cf. fig. 8) indicating no major changes in the deformation
structures in the grains during the performed cycling (C4-C7). By identifying the most intense high-intensity peaks in the high resolution reciprocal space maps from all six load steps (L2 and C4-C8), it was possible to follow four large subgrains through the performed 7350 tension-tension cycles corresponding to an accumulated strain of 6.7 – undeterred by the drop in beam intensity after loading step C8. The constant subgrain volume fraction and the fact that the very same subgrains were found after 7350 cycles indicate that changes in the deformation structure established during tension to 1.3% are rather minor despite the significant number of cycles and confirm the presence of quite stationary deformation structures during the preformed tension-tension cycles.

This is further supported by the behaviour of the radial profile of the entire grains. The peak position after cycling remains almost constant for each grain (cf. fig. 9) despite the fact that during an individual single cycle significant changes of the peak position occur (Diederichs et al. 2017a). As all measurements presented here were acquired after pausing the cyclic deformation at the point of maximal displacement, i.e. the upper reversion point of the hysteresis curve, similar peak positions are expected; the observed small, systematic shift of the mean radial position of each grain towards higher diffraction vectors is caused by stress relaxation during cycling (cf. section 4.1).

The integral width of the radial profiles decreases significantly after the first cycling step (C4), but stays nearly constant for all following cycling sequences (C5-C8) indicating very little structural reorganization as also evidenced by the azimuthal maps. In this respect, the integral width after the first 800 cycles after loading (C4) seems to differ for all four grains from the trends observed for the profiles during subsequent cycling up to 6650 further cycles. This may indicate that during the first cycling step after tensile loading reorganization occurs, where the deformation structure introduced by tensile loading adjusts slightly to accommodate cyclic deformation as described by Mughrabi et al. (1978) for pure copper. As the first HRRSM was acquired after already 800 cycles after tensile loading, this initial reordering cannot be analysed in detail.

Quantification of the changes during the first 800 cycles hinted towards only a minor adaption of the deformation structure to the new deformation condition. If any structural adaption has occurred, it may have already been completed then, as no obvious changes occur during the following cycling steps. An even longer cycling may have caused further changes, but complementary ex-situ investigations revealed, that significant changes after admissible number of load cycles in synchrotron experiments (i.e. several thousand cycles) appear only if the strain amplitude is sufficiently larger.
4.3 Interpretation in terms of composite model

Internal stresses due to the composite nature of the deformation structure in each grain determine the asymmetry of the grain profiles and seem not to change distinctly during tension-tension cycling with the selected parameters (cf. Fig. 11b), in contrast to uni-directional tensile deformation (cf. Fig. 11a). It was observed that the particular grain 1 with high initial asymmetry caused by pre-deformation shows only minor changes in asymmetry and eventually develops an asymmetry comparable to the others. Both the difference between the asymmetry of different grains and the spread in the position of the subgrains of an individual grain (cf. Fig. 11a) become less pronounced during cycling indicating a levelling of the internal elastic strains and a stabilizing of the deformation structure.

The absolute asymmetry of all grains is positive as expected for observations under tension for diffraction vectors along the tensile axis. Dislocation-free subgrains, which formed during tensile deformation, yield plastically at lower applied stresses than dislocation walls. According to the composite model (Mughrabi 1983, Pantleon et al. 2010), plastic deformation in the mechanically softer regions lead to accumulation of interface dislocation, which cause forward stresses in the dislocation walls and back stresses in the subgrain. These internal stresses are responsible for the observed elastic back strains in the subgrains and hence the positive asymmetry of the entire profile.

The peak positions of the individual resolved subgrains follow the changes in the applied macroscopic stresses similar to the peak position of the entire grain (cf. Fig. 13). The peak position of the subgrains compared to the position of the grain, i.e. the peak shift, show in average the presence of elastic back strains (cf. Fig. 14b) in full accordance to the classical composite model (Mughrabi 1983). Their individual peak positions follow a Gaussian distribution (cf. Fig. 12) centred around the position of the peak maximum as expected from the refined composite model (Pantleon et al. 2010). Additionally, a peculiar size effect was discovered based on a statistical analysis of the individual subgrain profile positions and their peak shifts. Larger subgrains experience larger internal back strains than smaller ones (cf. Fig. 15). This effect can be explained by presuming that larger subgrains have a lower yield stress than smaller ones as dislocations require lower resolved shear stresses to pass between obstacles of larger distances by bowing (Kuhlmann-Wilsdorf and Van der Merwe 1982) evidenced, for instance, by deformation structures with smaller average subgrain sizes having reportedly larger yield strengths (Staker and Holt 1972, Gil Sevillano et al. 1980, Raj and Pharr 1986). When applying increasing tensile stresses, large subgrains yield first and experience locally the largest plastic strain. Consequently, they develop the largest interface
dislocation density and hence the largest elastic back strains.

The discussed levelling of the internal stresses (cf. Fig. 14) leading to more similar values of the elastic back strain of different subgrains might be caused by concurrent work-hardening of individual subgrains harmonising the initial differences in yield strength between them. Yielding of dislocation walls around the largest subgrains due to the largest forward stresses will restrict the density of stored interface dislocations and hence limit any further increase of the elastic stresses. These ideas can explain the observations on the distributions of peak shifts of the subgrains (cf. Fig. 12): The initially large peak shifts of the largest subgrains remain unaltered with cycling, while the peak shifts of smaller subgrains continue to increase causing an increased average peak shift and a more uniform distribution with smaller spread.

5. Summary

High resolution reciprocal space maps of 400 diffraction peaks of four different grains were acquired in-situ after a number of different tensile loading steps and subsequent tension-tension cycles, while monitoring macroscopic axial stress and strain in-situ.

- Several individual subgrains within a selected grain are followed through 7350 tension-tension cycles corresponding to an accumulated strain of 6.7.
- Azimuthal maps and radial profiles do not change significantly during the cyclic deformation for the investigated number of tension-tension cycles (7350 in total). The radial peak profiles shift as a result of stress relaxation during cycling, while the peak width and positive asymmetry keep an almost constant value indicating neither major increase of dislocation density, nor major rearrangement of the microstructure during the investigated cycling.
- Changes in peak width and asymmetry during the first 800 cycles following the tensile loading step compared to that of subsequent cycling steps were observed which were attributed to an initial adjustment of the deformation structure from tensile deformation to the cyclic conditions.
- Individual dislocation-free subgrains were identified and their peak position was analysed. The majority of subgrains experienced elastic back strains though in different extent, following a Gaussian distribution. Changes in the distribution during cyclic deformation are attributed to a levelling of the internal stresses due to work-hardening in soft subgrains and plastic yielding of their surrounding dislocation walls.
- For all load steps, the largest subgrains retrieved in the analysed grains experienced the largest elastic back strains.
This is rationalized by a smaller yield stress of larger subgrains, leading to a larger plastic deformation and hence requiring the largest elastic back strains to maintain compatibility.

The investigations demonstrate that HRRSM is a suitable technique enabling microstructural analysis of grains and subgrains in-situ during cyclic deformation for thousands of cycles. By investigating the behaviour of individual subgrains the description of changes in the deformation structure in terms of the composite model is improved.

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Highlights

- Cyclic deformation is monitored in-situ by high resolution reciprocal space mapping
- Individual subgrains are followed through 7350 tension-tension cycles i.e. a strain of 6.7
- Peak width and profile asymmetry of grain profiles reveal adaption of deformation structure during the first cycles
- Elastic back strains of subgrains show peculiar size effect
- Gaussian distribution of the peak shift of subgrains evolves with cycling

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