Understanding the evolving state of deforming polycrystals using synchrotron x-rays

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Abstract. The multiscale nature of irreversible processes like plasticity make them particularly challenging. One must be looking everywhere, all the time to gain a predictive understanding of plasticity-related processes such as fatigue crack initiation. High energy (HE) synchrotron x-ray diffraction is a unique tool for characterizing the state of an entire aggregate of metallic crystals with sub-crystal resolution. Combined with \textit{in situ} loading, HE x-ray diffraction provides experimental hope for characterizing the conditions leading to the initiation of a crack. In this work, we conducted a HE x-ray diffraction experiment on a commercially pure copper sample subjected to \textit{in situ} cyclic loading at the F2 Station at the Cornell High Energy Synchrotron Source (CHESS). We observed that the evolution of lattice orientation distributions within each crystal evolved as the specimen was loaded through complete fatigue cycles. We defined a “size” of these orientation clouds, which we called $\Theta$. Changes in $\Theta$ are related to the plasticity-induced state of each crystal. We saw significant evolution of the $\Theta$ distributions as the sample went through cycle 2. By cycle 256, the evolution of the $\Theta$ distribution was significantly reduced - consistent with the saturation of cyclic hardening that we see on the macroscale. We also saw that the 40 crystals with the largest values of $\Theta$ in the first half cycle had a larger amount of $\Theta$ accumulation on average over the 256 cycles than the rest of the crystals in the aggregate.

1. Introduction

High energy (HE) synchrotron x-ray methods have significantly impacted the scope of the scientific questions we are able to explore. Working together with x-ray scientists and the manufacturers of sophisticated specimen interrogation tools and ever faster x-ray detectors, the structural metals community has transformed x-ray science experiments into a new generation of materials characterization measurements. HE x-ray diffraction experiments employing \textit{in situ} loading are particularly well-suited for studying loading history-dependent processes such as plasticity. Indicators of material state provided by traditional “before and after” 2D snapshots of material microstructure from a micrograph or an electron backscatter diffraction (EBSD) map are greatly augmented by the real-time measures of the distributions of lattice spacings (strains) and lattice orientations at the size scale of a polycrystalline aggregate and below that can be obtained using HE x-ray diffraction. This is particularly true for processes like fatigue crack initiation, which are among the most challenging to understand, model and ultimately predict.
because of their seemingly random nature. The ability to “track” the plasticity of each crystal during each cycle could lead to the understanding necessary to predict the initiation of a fatigue crack. A state feature (state variable) with subgrain scale resolution that would provide coverage over an aggregate is necessary; something that would mark grains that are more susceptible to crack initiation by virtue of the fact that they have incurred “more plasticity”. This paper describes a study of the evolution of an aggregate of commercially pure copper crystals subjected to cyclic loading conditions consistent with low cycle fatigue (LCF); each fatigue cycle contains macroscopically elastic-plastic deformation. The uniaxial test specimen initially loads elastically and yields in tension, then unloads elastically and re-yields in compression. The macroscopic cyclic stress-strain curve forms characteristic hysteresis loops that change in size (typically increase) with each cycle reflecting the plasticity-induced material evolution that is taking place within each crystal. It has been hypothesized that at some point, the to-and-fro plastic slip within one crystal or a group of crystals initiates a fatigue crack. Using HE x-ray diffraction we studied the cyclic plasticity within every deforming crystal by measuring how the lattice misorientation changes as the sample is loaded around the hysteresis loop.

2. Experiments and Results

X-ray diffraction experiments were performed at the F2 beamline at the Cornell High Energy Synchrotron Source (CHESS). The sample was electrical discharge machined from 6.35 mm diameter commercially pure copper rod. The square gauge region had a cross-section measuring 1 mm by 1 mm. After manufacturing, the sample was heat treated in a nitrogen environment at 750° F for two hours. A GE area detector was utilized with an x-ray energy of 61.332 keV and a sample to detector distance of about 850 mm. During this experiment, a total of five, 125 micron tall layers were scanned at each load step, such that the previously mapped volume was contained within the far-field data. The sample was deformed for a total of 256 tension-compression cycles at a fixed strain amplitude of 0.3%. Cycling of the sample was performed under displacement control and the macroscopic strain was monitored using digital image correlation (DIC). The displacement endpoints at the hysteresis loop tips were periodically adjusted to maintain a consistent strain amplitude based on the DIC data. Figure 1 shows the hysteresis loops at cycles 2 and 256 along with the load steps where diffraction measurements were made. The differences between the size of the loops is indicative of the cyclic loading–induced evolution (cyclic hardening) that is a characteristic of commercially pure copper.

![Figure 1](image_url)

**Figure 1.** Macroscopic stress-strain data from the commercially pure copper sample deformed *in situ* cyclically in uniaxial tension/compression. Cycles 2 and 256 are depicted. Load steps where HE x-ray diffraction experiments were performed are also shown as red circles. (Note: load step 1T is the same as load step 6C but 1C and 6T are different.)
2.1. Diffraction data

Diffraction experiments (far field) were performed at 11 points (load steps) around the stress-strain hysteresis loops. The first load step (1C) is at the hysteresis loop tip corresponding to the maximum tensile strain in the sample, 0.3%. Load step 2C roughly corresponds to the specimen yield point, then 3 load steps approximately evenly spaced in strain followed by load step 6C, which was taken at the minimum strain point. Load step 1T is the same as 6C and the process is repeated on the tensile-going half of the loop. Note that point 6T is different than 1C. At each load step, deformation of the sample was halted and the load on the specimen was reduced by approximately 10%. Data from the approximately 350 crystals within the aggregate were gathered by rotating the sample around the loading axis. Diffraction data were collected continuously on a GE area detector and separated into 0.25° bins. Full 360° rotations, which enable collection of all peaks associated with a crystal, were made possible using the RAMS loading system, which is resident at CHESS F2 [1].

We are interested in understanding the evolution of plastic straining in each deforming crystal by examining the evolution of intragrain lattice misorientations. In previous work, we have connected the azimuthal spread of each diffraction peak associated with a crystal as an indicator of misorientation [2, 3, 4, 5]. This work employs a different methodology. Instead of fitting each diffraction peak, the orientation spread within each grain is estimated though a forward projection approach by employing a virtual diffractometer [6]. The set of orientations associated with each grain can then be examined. The orientations within one crystal at the tensile and compressive ends of hysteresis loops 2 and 256 are shown in Figure 2. Rodrigues orientation space - parameterized with the Rodrigues vector, \( \vec{r} = n \tan \frac{\varphi}{2} \), where \( n \) is the rotation axis and \( \varphi \) is the rotation angle - is discretized in Figure 2 and each subvolume containing mapped detector intensity is shaded. Predicted locations of intensity on the detector for each discretized point (defining a unique orientation) in Rodrigues space are compared to the experimental detector images [7]. Changes between tension and compression in cycle 2 - the cloud actually shrinks when the sample plastically deforms in compression - and the increase in the size of the regions (orientation clouds) with little difference in tension as compared to compression in cycle 256 are visually evident. A simple measure to quantify the “size” of the cloud for each crystal at each load step can be defined as

\[
\Theta = \frac{2}{N} \left( \sum_{i=1}^{N} \tan^{-1} \left| w_i \right|_{\text{load step}} - \sum_{i=1}^{N} \tan^{-1} \left| w_i \right|_{\text{initial}} \right)
\]

where \( w_i = r_i - \bar{r} \) is the misorientation of each subvolume relative to the current average orientation of the grain, \( \bar{r} \). Figure 3 and Figure 4 depict the \( \Theta \) histograms at each load step in cycles 2 and 256.

3. Discussion

We associate \( \Theta \) with the “amount” of plasticity-induced lattice misorientation present within a particular crystal - \( \Theta \) is an indicator of the plastic state of the crystal and changes in \( \Theta \) from one load step to the next are indicative of the changes in that state due to plasticity-induced orientation evolution within a crystal. In the data, we see \( \Theta \) values getting larger and smaller - both are indicative of plastic deformation. By definition, the \( \Theta \) distributions shown in Figure 3 and Figure 4 are snapshots of the state of each and every crystal at each load step; the attributes of these histograms are related to the state of the aggregate. As can be seen, during cycle 2 there are significant changes in the histograms as the loading proceeds around the hysteresis loop. Beginning at the tensile loop tip (2-1C) the large positive tail on the distribution is seen to contract until by 2-5C it looks nearly Gaussian. It then spreads before the stress-strain curve hits the compressive loop tip (2-6C=2-1T). Evolution during the tensile-going part of the loop
Much more misorientation evolution is observed during cycle 2 when compared to cycle 256.

Figure 2. Orientation clouds within one crystal inside the deforming copper aggregate at the hysteresis loop tips during cycles 2 and 256. The clouds are depicted in Rodrigues orientation space.

Figure 3. Histograms of the misorientation metric, $\Theta$, for the crystals within the copper aggregate for cycle 2. (See Figure 1 for locations of the load steps.)

is somewhat similar, with the distribution actually narrowing from 2-1T to 2-2T and to 2-3T as the specimen re–yields in tension, then broadens again by 2-4T and eventually taking on a shape at 2-6T that is similar to but different than 1C, where the cycle started. The histograms in Figure 4 are much broader by cycle 256 at every point around the loop and the evolution of the histograms from one load step to the next is much more muted.
Figure 4. Histograms of the misorientation metric, $\bar{\Theta}$, for the crystals within the copper aggregate for cycle 256. (See Figure 1 for locations of the load steps.)

3.1. Upper Tail of the Initial $\bar{\Theta}$ Distribution
One of the most important developments from the recent HE x-ray diffraction research on polycrystalline metals is the ability to quantify how the crystal-scale stresses and strains—and underlying crystal states—vary from one crystal to another and how those crystal states evolve with plastic deformation. One crystal or set of crystals within the copper aggregate will eventually accumulate plastic slip, become more prone to crack initiation, and a crack will form. The overarching goal of this project is predicting that process. With the hypothesis that “more slip” will lead to crack initiation, the first crystals to examine as likely crack initiation sites are those experiencing the largest inelastic strains. An interesting set of crystals to track, therefore, are those that yield first and accumulate the most plastic strain during the first half cycle. The $\bar{\Theta}$ distributions at the hysteresis loop tips of cycle 1, (1-1C and 1-1T), are shown in Figure 5 along with the first loop tip of cycle 2 (2-1C). The $\bar{\Theta}$ distribution at the 1-1C load step is due to the initial loading; the crystals in the upper tail were those most susceptible to slip in the initial state of the specimen. This seems a likely set of crystals to track in terms of $\bar{\Theta}$, so we begin our search for the most fatigue vulnerable crystals by examining the 40 crystals with the largest $\bar{\Theta}$ values at point 1-1C. We call those crystals the Top 40. The rest of the crystals we label as Bulk. Figure 5 also depicts how the characteristic “long upper tail” in the 1C $\bar{\Theta}$ distributions (from both cycles 1 and 2) could actually be the superposition of 2 Gaussian distributions, the Top 40 and the Bulk.

At load step 1-1C, we identify the Top 40 and Bulk crystals then compute the mean $\bar{\Theta}$, $\bar{\Theta}_{avg}$, for both groups of crystals at the available load steps in cycles 1, 2 and 256. Since we are actually interested in how plastic slip increases in each crystal during a cycle, changes in $\bar{\Theta}_{avg}$ from one load step to the next and how those changes accumulate during a cycle are most interesting. We therefore define the sum of the absolute value of each increment of $\bar{\Theta}_{avg}$ from one load step to the next within a cycle by creating a quantity, $\bar{\Sigma}_L$, for load step $L$:

$$\bar{\Sigma}_L = \bar{\Theta}_{avg}^1 + \sum_{i=2}^{L} |\bar{\Theta}_{avg}^i - \bar{\Theta}_{avg}^{i-1}|$$

Values for $\bar{\Theta}_{avg}$ and $\bar{\Sigma}$ at each load step are given in Table 1 and $\bar{\Sigma}$ is plotted vs. load step in Figure 6.

From Figure 6 we see a much larger increase in $\bar{\Sigma}$ in the Top 40 crystals compared to the Bulk. The $\bar{\Sigma}$ curves become roughly parallel until a slope change in the Top 40 curve at 2-4T,
which corresponds to a reversal in the $\bar{\Theta}_{\text{avg}}$ data trend, which can be seen in Table 1. In terms of the underlying cloud data, this reversal means that, on average, the clouds stopped getting smaller and began getting larger again. There was also a reversal in $\bar{\Theta}_{\text{avg}}$ in the compressive-going data between 2-5C and 2-6C but this didn’t produce any noticeable slope change in the $\Sigma$ data. During cycle 256, the $\Sigma$ data remain roughly parallel - separated by nearly the same amount as at load step 2-6T taken 254 cycles earlier. Note, however, that this may not mean that the differences in the $\Sigma$ values that we see in cycle 256 represents all the changes in $\bar{\Theta}_{\text{avg}}$ that accumulated between cycles 2 and 256. Reversals of the change in $\bar{\Theta}_{\text{avg}}$ similar to what we saw in cycle 2 are most likely present in other cycles - especially early cycles - these reversals could produce additional increases in $\Sigma$.

Figure 5. (Left) Cycle 1 $\bar{\Theta}$ distributions at the hysteresis loop tips. (Right) Schematic depicting how the 1C $\bar{\Theta}$ histogram could be decomposed into 2 Gaussian distributions: the Top 40 crystals and the Bulk crystals.

Table 1. $\bar{\Theta}_{\text{avg}}$ and $\Sigma$ in degrees for the Top 40 and Bulk sets of crystals.

| Load Step | Cycle 1 | Cycle 2 | Cycle 256 | Cycle 256 |
|-----------|---------|---------|-----------|-----------|
|           | Top 40  | Bulk    | Top 40    | Bulk      | Top 40    | Bulk      |
|           | $\bar{\Theta}_{\text{avg}}$ | $\Sigma$ | $\bar{\Theta}_{\text{avg}}$ | $\Sigma$ | $\bar{\Theta}_{\text{avg}}$ | $\Sigma$ |
| 1C        | 0.0787  | 0.0787  | 0.0272    | 0.0272    | 0.1097    | 0.1894    |
| 2C        | -       | -       | -         | -         | 0.0826    | 0.2165    |
| 3C        | -       | -       | -         | -         | 0.0610    | 0.2381    |
| 4C        | -       | -       | -         | -         | 0.0492    | 0.2499    |
| 5C        | -       | -       | -         | -         | 0.0375    | 0.2616    |
| 6C = 1T   | 0.0389  | 0.1185  | 0.0342    | 0.0342    | 0.0445    | 0.2687    |
| 2T        | -       | -       | -         | -         | 0.0419    | 0.2714    |
| 3T        | -       | -       | -         | -         | 0.0459    | 0.2754    |
| 4T        | -       | -       | -         | -         | 0.0698    | 0.2993    |
| 5T        | -       | -       | -         | -         | 0.0993    | 0.3288    |
| 6T        | 0.1097  | 0.1894  | 0.0395    | 0.0395    | 0.1234    | 0.3529    |

3.2. Crystal Stresses, Strain Hardening and Crystal-Based Modeling
Of course plastic straining, in general, and the heterogeneity of plastic slip on the crystal scale, in particular, are intimately coupled to the stress experienced by the crystal as the sample is cycled.
In pure copper - due to its high degree of elastic anisotropy and complex dislocation interaction potential - the fully three-dimensional stress states at the crystal scale, even under uniaxial loading conditions, can be quite complicated and can evolve significantly during deformation. The high degree of cyclic strain hardening manifests on the macroscale as the differences in the cycle 2 and cycle 256 hysteresis loops presented in Figure 1. For a truly comprehensive understanding of the factors leading to fatigue crack initiation, lattice strains (and resulting crystal stress tensors) should be collected along with the orientation clouds. Unfortunately, due to the low strength of the pure copper, the lattice strains acquired during our experiment were very near the resolution level of the HE x-ray diffraction method - especially during cycle 2 - so no lattice strain or crystal stress data are presented. The question of how to bring the various aspects of the diffraction data - orientation clouds and crystals stresses, for instance - remains, however. The “best”, most rational use of the diffraction data for truly predicting fatigue crack initiation is in conjunction with a crystal-based deformation model. Comparisons between measured and simulated clouds and stresses can be made most readily. In simulations conducted in parallel with our experiments, the organization of crystals into persistent slip networks is proposed as an initiation mechanism [8]. We will explore this hypothesis in future work.

4. Summary, Conclusions and Future Work

We conducted HE x-ray diffraction experiments in situ during cyclic loading experiments on a polycrystalline sample containing over 300 commercially pure copper crystals. The focus of this work is on the plasticity–induced fatigue crack initiation that occurs in ductile metals such as copper; the to and fro plastic slip that occurs within a copper crystal during cycling has long been associated with crack initiation. The main goal of this particular study was to use the diffraction data and a virtual diffractometer employing a forward projection algorithm to approximate the evolution of the cloud of orientations within each crystal at load steps around

**Figure 6.** $\bar{\Sigma}$, the accumulated average $\bar{\Theta}$ within the Top 40 and Bulk crystals, at each load step around the hysteresis loops.
the hysteresis loops. Changes in the size and shape of the orientation cloud associated with a crystal indicate that plastic straining has occurred and, to a first approximation, the magnitude of the cloud change is related to the amount of accumulated plastic strain. We created a scalar measure of cloud size, $\bar{\Theta}$, to track the plastic state of the aggregate as loading proceeded around hysteresis loops of a $\pm 0.3\%$ uniaxial cyclic test. Histograms of $\Theta$ early in the life of the sample (cycles 1 and 2) and later (cycle 256) were presented and the average $\Theta$ value, $\bar{\Theta}_{\text{avg}}$, from the 40 crystals in the upper tail of the histogram from the first cycle were tracked over cycles 2 and 256. We saw significant evolution of the $\Theta$ distributions in cycle 2 and almost no evolution in cycle 256; evolution on the crystal scale mimics the cyclic hardening we see on the macroscale in the commercially pure copper. The magnitude of the changes in $\bar{\Theta}_{\text{avg}}$ from one load step to the next, $\Sigma$, was computed and plotted. The $\Sigma$ values for the Top 40 crystals began the experiment larger than the Bulk crystals and were larger at cycle 256 indicating that on average, the crystals that yielded and plastically deformed the most at the start of the experiment, ended the experiment having accumulated “more” plasticity than the other crystals in the aggregate.

We introduced $\bar{\Theta}$ and the Top 40 as a first step to studying crystals we feel might be most susceptible to accumulating plastic strain. The HE x-ray dataset is incredibly rich and we have the details of the cloud distributions, as well as the average orientation of every crystal and their locations. We are currently examining the individual crystal data to understand vulnerability to fatigue crack initiation on a crystal by crystal basis. We are examining orientation dependence on $\bar{\Theta}$ and the role that crystal topology plays. Are the Top 40 crystals somehow organized so that persistent slip networks form [8]? Also, since fatigue crack initiation is a surface phenomena, how many vulnerable grains are on the surface of the sample? We will build on the concept of the orientation clouds and especially $\bar{\Theta}$ in the future - along with incorporating simulation results - as we continue to create new ways to use HE x-ray data to create understanding of complex phenomena like fatigue crack initiation.

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