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Abstract: We have used the cross-correlation based high resolution electron backscattering diffraction (HR-EBSD) technique to evaluate at high spatial resolution the spatial patterning of the type III intragranular residual stresses and geometrically necessary dislocation (GND) density within polycrystalline Cu after cyclic deformation. Oxygen free high conductivity (OFHC) polycrystalline copper samples were cyclically deformed under stress-control at a load ratio of 0.1 and EBSD measurements were made at points throughout the early stages of fatigue when strain amplitudes and cyclic creep rates are changing most significantly, namely at 0 cycles, 2 cycles, 200 cycles and 2000 cycles. Statistical analysis is presented showing that moderate correlations exist between stored GND density, residual intragranular stress and distance from the nearest grain boundaries or/and triple junctions.

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Manuscript: "Evolution of intragranular stresses and dislocation densities during cyclic deformation of polycrystalline copper"

Dear Prof Mahajan

We are very pleased to submit our revised manuscript to Acta Materialia for considering publication. Each of referee's comment and suggestion has been carefully addressed. Detailed explanations are included in the 'Response to Reviewer' file. All changes and corrections to our original manuscript have been highlighted in the revised manuscript.

We would be very grateful for Editor's effort and time to process our manuscript and very look forward to hearing from you soon.

Yours Faithfully

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Evolution of intragranular stresses and dislocation densities during cyclic deformation of polycrystalline copper
Acta Materialia

Authors are very grateful to Reviewer's time, effort and constructive comments. In order to comprehensively revise our manuscript and give most appropriate response, we address Reviewer's comments one by one in detail in the following section. (Reviewer's comments are marked using red colour, correction to the manuscript are marked as yellow and Authors responses are marked as blue.

Editor's/Reviewer's comments:

This paper gives some interesting insights into dislocation and stress development during cyclic testing. Overall the analysis is appropriate, although I was surprised about two things: the correlations reported in the paper are pretty low; I was surprised that the authors did not pursue this a little further to factor out some of the issues (such as substructure development) that appear to play an important role in causing this. Relating to the previous point, the authors mention dislocation substructure development in an extremely cursory and handwaving fashion. They mention correlations with orientation and different types of structure, but in an almost off-handed way that does not even connect with the wealth of literature on the subject. Issue 2. should certainly be fixed, and it would be nice to improve issue 1. a little if possible. I have made comments on the manuscript that I hope will be helpful (including pointing out a few typos).

1. These coefficients are low. The correlation coefficients are generally not very high (~-0.2)
(That's a bit of a stretch)

This is a sensible suggestion and we have rephrased this sentence as:

The correlation coefficients are generally not high (~-0.2)

2. which we attribute to rather complex and multi-factored interactions involving both elastic and plastic anisotropy along with complex and variable grain shapes, micro-texture and subsurface structure which all combine to influence on their final GND density and residual stress distributions.

It seems like the substructure causes a lot of noise here, but even if you factored that out, presumably the GND and stress build-up near the GB is still not strong? I wonder whether the correlation is stronger closer to the boundary, but as I note on the plots, the way that they are plotted makes this harder to see.

The substructure should not really be viewed as 'noise' as it is a true aspect of the material's response rather than an unwanted artefact or error in the measurement. However, the Reviewer's point that this internal structure within the grains tends to reduce the correlation between GND density and stress with distance from the GBs does seem to be correct and is the reason for the correlation coefficients being lower than naïve initial expectation. As this substructure is not really 'noise' it is not clear how it could be factored out. We chose not to pre-bias our analysis to only consider near boundary regions. The 0.5 µm step size limits the precision with which the boundaries

are located and makes the distance data rather discrete at small distances. We feel these factors would limit the value of analysis based only on points close to boundaries. We have modified the text to try to capture the reviewers comment on substructure reducing the level of correlation.

which we attribute to rather complex and multi-factored interactions involving both elastic and plastic anisotropy which lead to marked internal dislocation substructure within the grains, along with complex and variable grain shapes, micro-texture and subsurface structure which all combine to influence on their final GND density and residual stress distributions.

3. Similar levels of correlation were found at all stages of cyclic deformation examined and also for the single load-unload 'tensile' sample. This may indicate that the GND dislocation structure developed in the first few cycles does not alter much in its general distribution during further cycling. Dislocation networks tend to be connected spanning across several grains and dislocation bands with high dislocation density bands running along grain boundaries as well as across grain interiors. Such longer range patterning is not obvious and lack quantification in most TEM studies. There is also some suggestion that the dislocation structure developed in a grain has some dependence on grain orientation.

I am very surprised that the authors did not tie this (albeit short) discussion into the literature - here or earlier (section 3). I'm not sure whether they mean Schmidt factor dependence or simply orientation dependence relative to the viewing surface. I would think some reference back to papers such as the following would be appropriate: Philosophical Magazine, Vol. 87, No. 33, 21 November 2007, 5189–5214 Philosophical Magazine, Vol. 87, No. 33, 21 November 2007, 5215–5235

This is a good suggestion and we have now added some discussion and references to studies linking dislocation substructure to grain orientation:

Such longer range patterning is not obvious and lack quantification in most TEM studies. However, similar to TEM studies and CPFE work, there is also some suggestion that the dislocation structure developed in a grain has some dependence on grain orientation. Orientation dependence of dislocation structure has been reported in the extensive TEM studies by Huang & Winther [Philosophical Magazine, Vol. 87, No. 33, 21 November 2007, 5189–5214 Philosophical Magazine, Vol. 87, No. 33, 21 November 2007, 5215–5235] for monotonic deformation in rolling and tension to larger strains than examined here. Recent HR-EBSD studies of GND structure formed in monotonically deformed copper seem to be in broad agreement and found that $\langle 011 \rangle$ and $\langle 001 \rangle$ oriented grains have more boundary-like dislocation structure and more regular dislocation cell structure is preferentially formed in $\langle 111 \rangle$ oriented grains [IJF 2015]. Gaudina and Feaugas [Acta Mater 2004 v52, 3097-3110] used TEM to document in detail the orientation dependence of dislocation structures found in 316L stainless steel after tension-tension fatigue.

4. A closer inspection on the GND maps and combining them with IPF maps especially for the 2 cycles and 200 cycles, it can be seen that the grains with $\langle 111 \rangle$ orientation with respect to the applied macro stress (in blue on the IPF map) tend to have relatively low GND density and regular dislocation cell structures, more irregular dislocation bands with significantly higher densities are formed in other grains. Evolution with increasing number of cycles of GND formed dislocation structure is not very strong however there does seem to be more patch-like patterns of dislocation bands in the 200 and 2000 cycle samples. A comparison between cyclic deformation and tensile deformation does not show obvious differences in the structure and density of stored GNDs.

This is actually not that obvious at first glance. It would be good to have some quantification here, or at least some better tie-in to studies of dislocation structures (see my comment on page 12). Is the structure intensity (e.g. contrast between GND density in and outside the bands) higher in certain orientations? Is it evolving? Is the size of the structure evolving? I'm not saying that this paper is necessarily about dislocation structure, but since the point is raised, there should be more discussion about it.

Our response to the previous point provides for the greater discussion of this observation as requested by the reviewer. Our recent publication (Int J Plas 2015 v69, 102-117) used HR-EBSD to examine GND patterning and its evolution during monotonic tension and was quantitative about aspects the review asks about here. The current work however sought to map elastic strain variations as well as GND densities which significantly slow the data collection. We are reluctant to over-interpret the data we have here and examining particular orientation components would lead to smaller data partitions than we would be happy with in some cases. We feel the qualitative observations are as far as we should go and have commented on grain orientation dependence and evolution with number of cycles. We have however added a sentence to mention the more quantitative analysis that has been undertaken for monotonic tension

A closer inspection on the GND maps and combining them with IPF maps especially for the 2 cycles and 200 cycles, it can be seen that the grains with $\langle 111 \rangle$ orientation with respect to the applied macro stress (in blue on the IPF map) tend to have relatively low GND density and regular dislocation cell structures, more irregular dislocation bands with significantly higher densities are formed in other grains. This agrees well with our more quantitative observations of much larger HR-EBSD maps of dislocation structures formed in monotonically deformed copper [Int J Plas 2015 v69, 102-117].

5. Evolution with increasing number of cycles of GND formed dislocation structure is not very strong however there does seem to be more patch-like patterns of dislocation bands in the 200 and 2000 cycle samples.

Again - I'm not sure what this means. A better link to previous literature might help tighten up the notation and discussion.

We have removed the phrase 'patch-like patterns' and reworded to keep terms consistent throughout this paper.

Evolution of GND formed dislocation structure with increasing number of cycles is not very strong however there do seem to be more dislocation bands of raised GND density formed in the 200 and 2000 cycle samples.

6. Elastic strain and residual stress can thus be precisely determined with elastic strain measurement sensitivity of $\sim 10^{-4}$ [32].

As far as I know, precise residual strains and stresses are not possible with this technique - only relative stresses and strains within a grain. Note - I see that this is cleared up at the end of the next paragraph, but the language should be more precise here.

Precise implies with high repeatability i.e. low scatter - but not necessarily true (i.e. accurate). However, we certainly do not wish to mislead on a very important point concerning the

measurement technique. We have changed the text to make it clear that strains (stresses) are relative to a reference point within each grain.

Elastic strain and residual stress variations relative to reference pattern selected within each grain can thus be precisely determined with elastic strain measurement sensitivity of $\sim 10^{-4}$ [32].

7. This allows 6 rather than only 3 constraints to be used in the GND density analysis [28, 39, 40].

The authors may wish to also reference a recent article with a more detailed look at this issue: [Ultramicroscopy133\(2013\)8–15](#)

The suggested paper is indeed very interesting. Authors have added it to our text.

This allows 6 rather than only 3 constraints to be used in our GND density analysis [28, 39, 40]. Other methods are also available to estimate GND density based on orientation gradients measured by EBSD [John wheeler] and [Ruggles and Fullwood 2013].

8. Figure 1, EBSD data on annealed copper sample (500 μ m x500 μ m). Larger than 10 °misorientation angle between two neighbouring points is defined as a grain boundary. (a) an inverse pole figure map plotted with respect to loading axis [100]. (b) Grain size distribution histogram showing the mean grain diameter is $\sim 10.3 \mu$ m (averaged by area). (c) an [100] inverse pole figure along loading axis showing weak texture (2x random).

Is some cleanup algorithm used here?

Copper samples were electrolytic-polished before EBSD mapping in order to obtain optimal pattern quality. This sample was in the annealed state and EBSD pattern quality was very high so that no clean up or artificial modification of the resulting orientation maps were needed or used here.

9. Figure 3, Grain morphologies and orientation maps combined with intragranular residual stress and GND density as a function of imposed number of fatigue cycles. (column wise from left): inverse pole figure maps plotted with respect to the loading axis (vertical direction along X2 axis), grain boundaries marked as black lines were defined as minimum misorientation of 10 degree between two neighbouring points; GND density maps in lines per m2. The white areas are unrecovered regions due to low EBSD quality or high misorientation between test points and reference point within that grain. The fourth to sixth columns correspond to three normalised intragranular residual stress component maps (σ_{11} , σ_{12}). Zero residual stress was assigned to the unrecovered white regions as shown in GND density maps.

Presumably this means fairly heavy cleanup in the IPF? This should at least be declared back in the methods section or something.

Although copper samples have been deformed plastically ($\sim 10\%$), great care was taken in sample preparation, and the patterns were captured unbinned, with low gain and long exposure compared to most EBSD analysis. As a result the EBSD software (OIM) did not have problem with pattern indexing. >95% of points were indexed confidently. We did not need to apply heavy clean-up in the IPF.

I can understand why this would give white areas in the stress maps, but not for the GND calculations. I must be missing something?

For the cross-correlation based HR-EBSD analysis, two key data quality parameters are used, namely: correlation peak height (PH) which indicate similarity between test and reference patterns and mean angular error (MAE) which describes the consistency of shifts measurement. If the measured below thresholds of PH or MAE, that point will be rejected from our analysis and show as an empty (white) points in our map.

Importantly both stress and GND density maps are derived from HR-EBSD cross-correlation pattern displacement measurement, and so both maps are affected in the same way.

We have added a sentence into the methods section to state explicitly that the rotations from the HR-EBSD analysis are used in the GND calculation.

This allows 6 rather than only 3 constraints to be used in the GND density analysis [28, 39, 40]. The lattice rotations used in the GND density calculations are taken from the HR-EBSD measurement rather than the Hough-based measurements as this provides considerably better sensitivity.

10. Figure 8, the top maps (a), (b) and (c) showing Euclidian distance to GBs in μm (a) and the statistical study of the relationships of Euclidian distance to GBs with GND density and absolute in plane shear stress (c) in 2000 cycles fatigued copper sample. Similarly the bottom maps (d), (e) and (f) showing Euclidian distance to triple junctions (d), quantitative analysis of the correlation of Euclidian distance to triple junctions with GND density (e) and absolute in-plane shear stress (f) in the 2000 cycles sample. Dataset was put into 40 bins in both x and y axes. Grey background intensity shows the number of data points in each bin on log2 scale. The coloured lines show the median and 5 and 95 percentile binned values with respect to y axis. Minimum 200 points were required to plot percentiles. The Pearson correlation coefficients were calculated based on all measured points.

FIGURE 8

I have gone back between this graph and the text description and I still don't understand what is being represented. There are no axis labels. I think I probably understand the plot, but I'm not sure.

We have modified the figure caption to try to make this clearer:

Figure 8: Data for 2000 cycle sample (similar plots for the other samples are given in the supplementary figures). Maps with colours showing the distance (in μm) from each pixel to the nearest grain boundary (a), or triple junction (d). The remaining plots examine the correlation between position in the microstructure on the vertical axis [represented by distance to nearest grain boundary (b and c) or distance to nearest triple junction (e and f)] and either GND density (b and e) or maximum in plane shear stress (c and f). The grey background intensity shows the number of data points at the corresponding range of distances and GND density or stress. The coloured lines show the median and 5 and 95 percentile binned values of distance with respect to values of GND density or stress along the horizontal axes. Minimum 200 points were required to plot percentiles. The Pearson correlation coefficients were calculated based on all measured points

We have also added to the text.

in the EBSD map, and YGB corresponds to the coordinates of all grain boundary points as shown in Figure 8 (a). Points that were closer to the edges of the map than to the nearest grain boundary

were excluded from the analysis. Figure 8(a) shows 2D colour map with each point represent its Euclidean distance to its nearest GBs. GB points are marked as blue points which are at the same position as GBs observed in 2000 cycles IPF plot in Figure 3. A similar analytical routine was used to calculate Euclidian distance to triple junctions as shown in Figure 8 (d).

FIGURE 8

These are extremely counter intuitive to me (to have the distance on the y-axis and the measured value on the x-axis). For example in (c) you get the full range of stresses at distance 0.3 for the bottom 5%, which may help appreciate the variance or noise in stress measurements, but not really how the stress changes with distance. Furthermore, I would expect that correlations would be higher closer to the GB when the structure internal to the grain is ignored, but that is harder to pick out the way the plots are drawn.

In fact all the quantities distance, GND density and stress are measured; so we have choice on which way to present the results. The questions we sought to address were where do the 'hot spots' i.e. larger values of GND density and/or stress tend to be relative to grain boundaries and triple junctions. This leads us to plot the data as distance from GB/TJ as a function of GND density or stress. As mentioned in point 2 above the 0.5 μm step size limits the precision with which the boundaries are located and makes the distance data rather discrete at small distances which would limit the value of analysis based only on points close to boundaries.

11. [29] Wilkinson.Angus TE, Vilalta-Clemente.Arantxa, Jiang.Jun, Britton.T.Benjamin, Collins. David M. Applied Physics Letters 2014;L14-07295

This doesn't seem correct

This has been corrected.

Evolution of intragranular stresses and dislocation densities during cyclic deformation of polycrystalline copper

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Abstract

We have used the cross-correlation based high resolution electron backscattering diffraction (HR-EBSD) technique to evaluate at high spatial resolution the spatial patterning of the type III intragranular residual stresses and geometrically necessary dislocation (GND) density within polycrystalline Cu after cyclic deformation. Oxygen free high conductivity (OFHC) polycrystalline copper samples were cyclically deformed under stress-control at a load ratio of 0.1 and EBSD measurements were made at points throughout the early stages of fatigue when strain amplitudes and cyclic creep rates are changing most significantly, namely at 0 cycles, 2 cycles, 200 cycles and 2000 cycles. Statistical analysis is presented showing that moderate correlations exist between stored GND density, residual intragranular stress and distance from the nearest grain boundaries or/and triple junctions.

Keywords

Fatigue deformation; geometrically necessary dislocations (GNDs); intragranular stress; High resolution electron backscattering diffraction (HR-EBSD);

1. Introduction

Research on fatigue failure has a long history with perhaps the earliest report given in 1837 by Wilhelm Albert who studied iron chain subjected to repeated loading [1]. It has been stated that fatigue accounts for at least 90 percent of all service failure due to mechanical causes [2], and that the cost of fatigue failure is high (4% of US gross national product [3]) and can lead to catastrophic incidents. Many studies have been carried out over the past ~150 years to improve our understanding as this is one of the most critical materials failure modes.

Significant progresses of fatigue deformation development especially fatigue crack formation have been made on single crystalline materials by characterizing dislocation slip, interaction and accumulation using transmission electron microscopy (TEM) [4-9]. These provide significant insight into very local mechanisms but necessarily avoid any effects of constraint imposed by neighbour grains in a polycrystal. The study of single crystals is an important building block but the majority of engineering structural materials e.g. metal alloys, ceramics and natural rock consist of aggregates of grains with characteristic crystallographic orientations.

Design and safety cases typically use physical insight from single crystals to inform empirical equations for life and failure tolerance and detailed microstructural variation is not taken in to account. This drives a safety culture which is very conservative and limits cost savings. Movement towards physically motivated design rules, informed from accurate and high fidelity microstructural investigations, will enable a transformation towards 'sufficient but not excessive' design, enabling improved life and a reduction in material used within components, thereby imparting significant savings in, for example, fuel consumption and CO₂ emissions in transport applications.

Extension of knowledge towards polycrystalline materials is constrained by limited supporting experimental evidence. In polycrystals the complexity of microstructure (combination of grain size and orientations) gives rise to heterogeneous deformation distribution. Therefore observations must not only consider the macroscopic loading conditions, such as an externally applied component level stress, in combination with stress risers such as structural defects like notches, but also consider microstructural effects on the distribution of cyclic slip and stress variations. Where 'weakest-link' failure modes exist, such as in fatigue, the local combination of grains shapes and orientations may elevate the likelihood of failure. To understand this, high spatial resolution characterisation techniques, with high fidelity, must be employed in order to resolve the inhomogeneous dislocation density, stress or strain distributions within grains and also across large areas to map sufficient number of grains to understand how aggregates of grains deform together to accommodate cyclic deformation.

Traditionally this problem has been tackled using either high spatial resolution techniques, such as TEM, or volume averaged high fidelity techniques such as high energy X-ray diffraction or high energy neutron diffraction. Diffraction contrast TEM observations have been highly influential in characterising dislocation sub-structures such as dislocation veins, persistent slip bands (PSBs), labyrinths, and cell structures that form during cyclic deformation [4, 8, 9]. However, the observed region of interest using TEM is limited to $\sim 10 \times 10 \mu\text{m}^2$ in a thin foil, which often only contain limited number of grains (potentially less than one in many cases) so that co-operative processes leading to dislocation patterning which spans many grains are typically not captured. High energy X-ray and neutron diffraction have mainly been used to probe macro residual stress (type I) distributions near large-scale component features such as holes and notches using diffraction peak positions [10]. Furthermore peak broadening occurs due to very short range stress fluctuations (type III) caused by dislocations introduced by plastic deformation and has extensive literature associated with it [11-14]. Although short range stress fluctuations are responsible for the effect only the average response over a relatively large volume is obtained and the true spatial distribution cannot be recovered. Innovations in synchrotron-based X-ray diffraction methods have provided routes to measure grain averaged residual stresses (type II) [15-17]. Transmission powder diffraction geometry is used with the sample rotated to generate patterns in which individual diffraction spots can be segmented and assigned to individual grains. A large number of grains are probed in parallel so statistical analysis can be undertaken but the spatial information is of limited resolution. New approaches, such as the reflection micro-Laue geometry, enables probing 3D strain elastic strain tensors (ie stress) stored within individual grains using a fine focused white beam and a differential aperture to distinguish the depth of different contributions to the observed patterns [18, 19]. While this technique is clearly an exciting innovation, sampling a large number of grains with sufficient spatial information in order to address fatigue deformation problem in polycrystalline materials remains a challenge.

One recent development used high spatial resolution (sub-micro) digital image correlation (HR-DIC) technique to correlate microstructure with surface plastic strain distribution during the fatigue crack formation process [20]. However, typically HR-DIC only captures in-plane total strains, and cannot easily track the driving stresses, out of plane strain and residual deformation such as lattice curvature and residual stress. Many of these studies have been largely qualitative in nature [21] and complement analysis of out of plane slip through slip trace analysis [22].

In contrast, the residual elastic strain state is now accessible with EBSD technique. Wilkinson et al in 2006 [23] developed image cross-correlation based high resolution EBSD technique by measuring the shifts of test EBSD patterns with respect to a reference EBSD pattern within a grain. This technique significantly improved angular resolution of conventional EBSD technique and provided full 3D elastic strain and rotation tensors (with appropriate assumptions). When combined with the existing advantages of EBSD including fine spatial resolution ($\sim 40\text{nm}$) [24], potential to scan large areas, easy access to complementary SEM imaging modes (e.g. electron channelling contrast image (ECCI) [25] and cathodoluminescence (CL) [26]), and the improvement in angular resolution makes HR-EBSD a powerful technique to study fatigue deformation process in polycrystalline materials. Considerable development of the method has been made such that the analysis generates rather comprehensive deformation information such as intragranular residual stresses (type III) [27], geometrically necessary dislocation (GND) density [28] and total dislocation density [29]. These quantitative measurements can be readily combined with the microstructural information more commonly available from 'conventional' EBSD such as grain orientations, phase distributions, grain size, proximity to grain boundaries and triple junctions to reveal microstructurally sensitive trends.

The aim of this study is to systematically characterize the distribution and evolution of residual stresses, GND density and total dislocation density in FCC polycrystalline copper samples at the early stages of tension-tension fatigue using the HR-EBSD technique. Our study combines qualitative visual inspections of the residual stress and GND density maps and the patterns formed with respect to microstructural features along with quantitative analysis of statistical distribution and development with increasing number of cycles. Furthermore, samples subjected to cyclic and monotonic loading up to approximately the same strain will be compared.

2. Experimental and analytical methods

Four dog-bone-shaped OFHC (99.95% purity) copper specimens were milled from a 2 mm thick copper sheet. These specimens were progressively ground and polished to a $1\mu\text{m}$ finish. Colloidal silica was then used to give a final mechanical and chemical polish. These polished specimens were annealed at 550°C for 15 minutes and furnace cooled to the room temperature to reduce residual cold work from the manufacture process. Finally electrolytic-polishing in 85% phosphoric acid solution was utilised to remove the oxidation layers and contamination formed on the free surface to give optimised results for EBSD measurements.

A preliminary EBSD map (>1000 grains) of the annealed copper was collected to reveal the general microstructure morphology and orientation distribution as shown in Figure 1 (a). The average grain size is determined as $\sim 10.3\mu\text{m}$ (Figure 1 (b)) and there is a relatively weak texture existing in the annealed samples as indicated in Figure 1 (c).

Stress controlled fatigue tests with a stress range of $\sim 150\text{MPa}$ ($16.6\text{-}166\text{MPa}$) at load ratio of 0.1 were performed at a frequency of 1Hz on these specimens to increasing number of cycles (0 cycle, 2 cycles, 200 cycles and 2000 cycles) as illustrated in Figure 2(a). Interpolation of life time data presented by Murphy [30] indicates that these loading conditions should correspond to fatigue lifetimes of $\sim 10^6$ cycles. The 0 cycles sample provides a reference annealed state and 2 cycles sample shows the initial deformation state at the start of fatigue deformation process. Significant cyclic hardening has been imposed onto the 2 cycles sample as illustrated by the change of cyclic loop width in Figure 2(b) and also as manifested by the gradient of cyclic creep strain as shown in Figure 2(c). At 200 cycles, the sample has reached a stable cyclic state where further cyclic hardening is not obvious as shown in Figure 2(b). Nevertheless cyclic creep strain continues to increase at constant rate as a function of number of fatigue cycle. A sample subjected to 2000 cycles fatigue was also selected as many small intragranular fatigue cracks had been formed on free surface as shown in Figure 2 (d). To compare with monotonic deformation, a copper sample was chosen to be deformed monotonically to 10% strain which has been reported in another study [27]. The central gauge section of each specimen was cut out using a slow saw such that the induced cutting deformation can be minimised.

EBSD measurements were carried out using a TSL OIM EBSD system on the central region of these gauge sections in a JEOL 6500F SEM. An electron beam with 20kV acceleration voltage and $\sim 16\text{nA}$ current were used to scan EBSD maps of a size of $113\mu\text{m} \times 113\mu\text{m}$ and a step size of $0.5\mu\text{m}$. This step size was selected based upon prior work [31]. All EBSD patterns were saved at their optimum quality (zero gain, no contrast adjustment, 1x1 binning and ~ 1 second exposure time) into 1000 pixel x 1000 pixel 12 bit depth tiff images during the scanning process such that optimum angular resolution in cross-correlation process can be achieved. Elastic strain and residual stress variations relative to reference pattern selected within each grain can thus be precisely determined with elastic strain measurement sensitivity of $\sim 10^{-4}$ [32]. The mapped regions were selected at random near central region of each sample.

The crystal orientations were determined using OIM DC and microstructure maps including grains and grain boundaries with a tolerance of 10° were calculated using OIM A v5. These were imported into an in-house high resolution EBSD analysis tool [23, 33-36]. Reference points for the cross correlation analysis were selected within each grain by finding points with the highest pattern quality towards the grain interior; it is not necessary, and is highly unlikely, that these reference points are strain free. The EBSD pattern shifts between the reference pattern and the remaining patterns obtained from within each grain were measured using the cross-correlation method [37]. Remapping was applied by removing the rotation determined from the initial cross-correlation pass in order to minimise artefacts induced in the elastic strain by large rotation [35]. Eight of nine components of the displacement gradient tensor can be determined in this way, namely three misorientations, three shear strains and then two terms which describe differences in the normal strain components. The ninth component is the volume change which alters the Kikuchi band widths but does not lead to any shifts in the band positions. This final degree of freedom, the hydrostatic strain is found using the boundary condition that the normal stress across the free surface is forced to be zero using knowledge of the single crystal elastic constants (more detail of the analysis route can be found in [33]). After determining elastic strain variations from the unknown reference point strain state Hooke's law was used to find the corresponding stress variations. The mean stress within each grain was then calculated and subtracted for each point within a grain. In this way the residual stresses reported throughout the paper are the type III stress variations away from the mean stress in a particular grain.

Polar decomposition was used to partition the deformation gradient into a finite rotation matrix and an elastic strain tensor. According to Nye's theory [38], the GND density can be estimated from elastic strain and rotation gradients. The EBSD technique probes only the surface of a sample and therefore the gradients along the third direction (normal to the free surface) are not accessible. From our experimental results, it was found the magnitude of elastic strains were significantly smaller than the magnitude of lattice rotation. Under these circumstances, the elastic strain gradients can be neglected in the GND density determination so that Nye's analysis can be used rather than Kroner's extension which includes the elastic strain gradient terms but is less good at constraining the problem. This allows 6 rather than only 3 constraints to be used in our GND density analysis [28, 39 and 40]. The lattice rotations used in the GND density calculations are taken from the HR-EBSD measurement rather than the Hough-based measurements as this provides considerably better sensitivity. Other methods are also available to estimate GND density based on orientation gradients measured by EBSD [39, 40].

[28, 41, 42].

We link rotation gradients to stored GND density by assuming that in FCC crystals there are 12 possible types of pure edge dislocations and 6 types of pure screw dislocations. Solving for (up to) 18 different dislocation densities with only six lattice curvatures leads to a mathematically underdetermined problem. Therefore a physically motivated minimization scheme is used to find an appropriate set of dislocations with minimum dislocation line energy that is likely not unique but necessarily supports the measured lattice curvatures. The detailed mathematical framework of determination of GND density can be found in [28, 41, 42].

The total dislocation density, i.e. both geometrically necessary and statistically stored dislocations, for each map was estimated from the tails of residual stress probability distributions $P(\sigma)$ which extend to very high stress levels. Here the form of these extremes of stress can be linked directly to the probability of sampling near a dislocation core from which a probability distribution of the form $P(\sigma) \propto \frac{1}{\sigma^3}$ is anticipated. It has been demonstrated that the extreme stress values are consistent with this functional form [43]. The analysis follows from the framework developed by Groma et al for X-ray line profile analysis [43]. We use the method described by Wilkinson et al [44], who adopted Groma's approach for use with HR-EBSD measurements of residual elastic stress, and linked the restricted second moment of stress probability distributions within grains to the total dislocation density. Detailed mathematical framework can be found in the appendix. It should be noted that in this approach only edge dislocations with dislocation line directions normal to free surface are assumed to be present. The approach has been validated directly against discrete dislocation modelling and further demonstrated experimentally on epitaxial semiconductor layers, Cu tensile test pieces and cold rolled steels covering dislocation densities from 10^{13} to 10^{16} m^{-2} [44].

3. Results

3.1 Maps of microstructure, GND density and residual stresses

The distribution and evolution of the intragranular residual stresses and GND density with increasing number of repeated cycles are shown in Figure 3. In the annealed sample, the magnitude of GND density and residual stresses are very low. GND density increases significantly when comparing the annealed and deformed samples.

Comparing GND density in samples with increasing number of cycles, the overall magnitude of GND density does not vary significantly. In these deformed samples, very complicated GND networks are formed in which dislocation bands span across a number of grains and join up at some triple junctions and twin boundaries. A closer inspection on the GND maps and combining them with IPF maps especially for the 2 cycles and 200 cycles, it can be seen that the grains with $\langle 111 \rangle$ orientation with respect to the applied macro stress (in blue on the IPF map) tend to have relatively low GND density and regular dislocation cell structures, more irregular dislocation bands with significantly higher densities are formed in other grains. This agrees well with our more quantitative observations of much larger HR-EBSD maps of dislocation structures formed in monotonically deformed copper [45] Evolution with increasing number of cycles of GND formed dislocation structure is not very strong however there does seem to be more dislocation bands of raised GND density in the 200 and 2000 cycle samples. A comparison between cyclic deformation and tensile deformation does not show obvious differences in the structure and density of stored GNDs.

Three grain-normalized in-plane intragranular residual stress component (i.e. variation of the measured stress at each point within the grain from the grain averaged stress value) are shown in the fourth, fifth and sixth columns of Figure 3. Once samples were cyclically deformed (Figure 3, 2C), significant residual stress variation can be observed within grains and this is inhomogeneously distributed. Visual inspection suggests that relative low residual stress variation can be found in grain interiors and large residual stress variations tend to accumulate near grain boundaries and the sharp interfaces of twin boundaries. Significantly, the intragranular residual stress tends to relax with further increasing number of cycles with similar intragranular patterning.

Quantitative assessment of frequency histograms constructed using all valid measurement points within each of these maps are presented for GND density in Figure 4 and stresses in Figure 5. As shown in Figure 4, the general GND density distributions for all five datasets seem to follow approximately log-normal distributions. There is a significant increase in stored GND density once cycling starts, but subsequent cycling only results in subtle changes in the distributions, particularly the development of a tail in the distribution at high GND density. Comparison of monotonic deformation and fatigue again only shows subtle differences.

Quantitative analysis using frequency histograms of the type III intragranular stresses shows more significant variations in Figure 5. The use of a log-scale for the probability axis very strongly emphasises the low probability high stress regions of the distributions. As seen in Figure 3, normal in-plane stresses (σ_{11} and σ_{22}) have wider range of stress distribution than in-plane shear stress (σ_{12}). Very large residual stresses are generated after 2 cycles and then gradually relax with increasing number of cycles. This systematic stress relaxation trend is obvious in the in-plane normal stress histograms in Figure 5 (a) and (b). Eventually, in 2000 cycles sample the stress is relaxed to a distribution close to that for the 10% monotonically deformed sample. Similar sample to sample trends are seen for the in plane shear stress distribution (figure 5c).

Probability plots shown in Figure 6 (a) reveal that the central regions (i.e. low stress) can be described with a Gaussian (Normal) distribution. For 2 cycles, more than 70% of entire dataset is following a Gaussian probability function and a larger fraction does so for the other datasets. The gradient of the linear parts of these probability plots corresponds to the width of the Gaussian and this varies systematically with the number of cycles. Deviation of these distributions from Gaussian can be seen towards the ‘tails’ of the distributions.

These tails have a $P(\sigma) \propto \frac{1}{\sigma^3}$ form so that a plot of the restricted second moment $v_2 = \int_{-\sigma}^{+\sigma} P \sigma^2 d\sigma$ against $\ln(\sigma)$ gives a straight line (figure 6) the gradient of which can be used to estimate the total dislocation density (see [44] and the appendix). The estimated total dislocation density variations with increasing number of fatigue cycles is shown as a blue line in Figure 7. The total density of dislocations increases significantly at the first two cycles. This rapid increase trend is followed by a small drop of total dislocation density with increasing number of cycles from 2 cycles to 200 cycles. It then becomes relatively stable with small fluctuation between 200 cycles and 2000 cycles.

Comparison of total dislocation density and map averaged GND density shown in Figure 7 reveals that variations with number of cycles in the total dislocation density follow a similar trend but at higher density than variations in GND density calculated for a 0.5 μm patch length. For the cyclically deformed samples the GND density contributes approximately $\sim 1/8$ of the total dislocation density which is a markedly smaller fraction than the $\sim 1/2$ contribution for the monotonically deformed sample, or $\sim 2/3$ contribution for the annealed case. The standard deviation of the log-normal of GND densities increases appreciably from 2 cycles to 200 cycles as indicated by the error bars in figure 7.

Furthermore, as shown in Figure 7, the total dislocation density stored in 10% monotonically strained sample is significantly lower than 2 cycles fatigued sample, though with increasing number of cycles, the difference becomes smaller and it is very close to the value measured from 200 and 2000 cycles sample. The magnitude and range of GND density in the monotonically deformed sample is slighter lower but very close to cyclically deformed samples.

3.2 Hot spot identification

Visual inspection of Figure 3 suggested that accumulation of high GND density and high stresses are associated with microstructural features especially areas adjacent to grain boundaries and triple junctions. Here we further quantitatively examine the relationships of dislocation density and stresses with these two microstructural features by calculating the Euclidian distance to grain boundaries and triple junctions and quantify the correlation strength.

We examine the stress and GND density distribution point by point in the same area of 2000 cycles fatigued sample as shown in the fourth row of Figure 3. The magnitude of stress tensor is represented by the maximum in plane shear stress, such that a single scalar can be used.

For each point within the maps, the minimum Euclidian distance to its nearest grain boundary is determined according to:

$$\text{Euclidian distance } (i,j) = \text{Min}(\sqrt{(X_i - X_{GB})^2 + (Y_i - Y_{GB})^2}) \quad \text{Equation 1}$$

where the X_i and Y_i are the coordinates of the testing point in the EBSD map, X_{GB} and Y_{GB} corresponds to the coordinates of all grain boundary points as shown in Figure 8 (a). Points that were closer to the edges of the map than to the nearest grain boundary were excluded from the analysis. Figure 8(a) shows 2D colour map with each point represent its Euclidean distance to its nearest GBs. GB points are marked as blue points which are at the same position as GBs observed in 2000 cycles IPF plot in Figure 3. A similar analytical routine was used to calculate Euclidian distance to triple junctions as shown in Figure 8 (d).

Relationships between GND density, Euclidian distance, and maximum in-plane shear stress have been assessed through the generation of 2D frequency histograms, where the number of points with, for example, a specific range of in-plane shear stress and Euclidian distance to grain boundary is illustrated as an $\log_2(\text{intensity})$ in Figure 8 (b-c, e-f).

To help visualise these distributions, the top 5%, 50%, and 95% percentile values, of distribution of locations for particular ranges of GND density and in plane shear stress, are plotted and shown as green, red, and blue lines overlaid on the grey plot. These plots confirm that high GND density and high residual stress are preferentially accumulated near grain boundaries. The Pearson correlation coefficients between Euclidian distance to grain boundaries, GND density and stress are -0.24 and -0.19 respectively indicating not only that moderate correlations (range from 1 or -1 to 0) linking hot GND density and stress with close distance to grain boundaries, but also the GND density hot spots are slightly more likely to be found adjacent to grain boundaries than the stress hot spots.

Compared to the Euclidian distance to grain boundary, the Euclidian distance to triple junction again shows moderate correlations with GND density (Pearson correlation coefficient of -0.22) and residual maximum in plane shear stress (Pearson correlation coefficient of -0.19). Some, but not all, regions near triple junctions also tend to assemble high density GNDs and large stresses. It is clear from figure 8 that points with low stress or low dislocation density are found over a large range of distances from the nearest grain boundary or triple junction. This means that 'cold spots' are distributed both through grain interiors and near grain edges. At high GND density or high stress the lines for 5%, 50% and 95% all shift to smaller distances but the 50% and 95% lines move much more significantly, so that the 'hot spots' tend to be located at closer to grain boundaries or triple junctions and only rarely occur in the grain interiors. Figure 9 shows that there is also moderate correlation between the GND density and in plane maximum shear stress (Pearson correlation coefficient of 0.29). This suggests that GND density and stress hot spots are co-located.

The other macroscopic deformation states show broadly similar patterns of correlation between GND density, stress and position within the microstructure (as seen in Table1).

4. Discussion

As with the majority of annealed polycrystals the Cu samples showed considerable cyclic hardening in the very early stages of fatigue testing. Figure 2b shows that the width of hysteresis loops decreases drastically over the first ten or so cycles and then saturates at strain amplitude of $\sim 2 \times 10^{-5}$. The majority of the literature on cyclic hardening uses either (plastic) strain control or 'ramp' testing [46] rather than load control which is more common in fatigue life testing however our results are in reasonable accord with extensive strain controlled testing at zero mean stress by Lukas & Klesnil [47]. Similarly the cyclic creep rate also decreases into a steady state condition during the first few cycles after which the steady state cyclic creep rate is approximately consistent with extrapolation from the higher stress tests reported by Kwofie [48].

Cross-correlation based HR-EBSD enables intragranular residual stresses, GND density and total dislocation density to be estimated from obtained EBSD patterns which substantially extends the deformation characterization capability on microstructural scale. This has been combined with conventional Hough-based EBSD to explore how materials behave as a result of applied cyclic stress at local scale in OFHC polycrystalline copper. This enables high fidelity, high resolution, and quantitative assessment across many grains and provides very rich quantitative data sets. Perhaps the most striking observation is that the magnitude of the residual stress variations within the grains was high after the initial large plastic strains of the first load-unload cycles but then diminished continuously over the next few thousand cycles (fig 5). This is in marked contrast to the relatively unchanging GND and total dislocation density (fig 7) in the fatigued samples. Biermann et al [49] made X-ray diffraction measurements from single grains of Cu polycrystals unloaded from various points around a stable stress-strain loop established by cycling for 2000 cycles under plastic strain control at an amplitude of 2.5×10^{-3} . Asymmetry in the $\{200\}$ peak profiles was greatest towards the extremes of the stress-strain loop and was used to evaluate opposing internal stresses in cell interiors and walls differing by 30-25 MPa as averaged over the sampled grain. The EBSD measurements presented here of course show much greater variation as they arise from much smaller volumes of material ($40 \times 120 \times 20 \text{ nm}^3$). Figure 9 shows that GND density and type III residual stress level are correlated. However, the separation of the data into cell interior and cell wall contributions is not as clear as it appears to be for the X-ray case where no requirement or means of checking the spatial consistency of the partitioning of the lattice strain data can be made. Biermann et al also used peak breadth analysis to imply dislocations densities of $1.3\text{-}4.2 \times 10^{14} \text{ m}^{-2}$ which are a little lower but in reasonable accord with values we have obtained.

Lorentzen et al [50] have used in situ neutron diffraction to measure average lattice strain responses for different grain orientation families during the first 8 cycles of deformation at total strain amplitude of 4×10^{-3} of an austenitic stainless steel. They showed amongst other things that the axial residual stress in the (002) grain family increased over the first ~ 5 cycles before showing a slight decrease over the next 3 cycles. Similar in situ experiments by Korsunsky et al [51] on 316L stainless steel report a decrease in the lattice strains at peak loads for testing beyond 10-50 cycles but did not report on residual stresses. Such measurements probe the mean grain averaged stresses but the EBSD measurements in Figure 7 show that the intragranular residual stresses also decrease over the first 2000 cycles. Wang et al [17, 52] used post-mortem neutron diffraction on 316LN stainless steel cycled under load control to show the average residual stresses in the (002) grain family decrease beyond ~ 5000 cycles to failure at ~ 8000 cycles, over which period peak breadth measurements implied dislocation densities of $1.4\text{-}2.2 \times 10^{14} \text{ m}^{-2}$. Recent high energy X-ray diffraction work reported by Obstalecki et al [53] shows the tantalising prospect of following the lattice strain tensor and peak broadening for individual grains within a polycrystal during in situ cyclic deformation. The measurements were made on a precipitation harden Cu alloy but loading was only carried out for two cycles at each of two strain amplitudes.

In general our GND formed dislocation networks agree with previous TEM characterization studies of fatigued copper at room temperature [6, 54, 55] in which dislocation cells and bands were commonly observed dislocation patterns; once dislocation cells were formed in the first few cycles, they would not change largely in the subsequent cycles and little difference in dislocation structure was found between monotonic and cyclic deformation. It is impossible to trace directly the evolution of particular GND dislocation structures and intragranular residual stresses with respect to microstructure features because four different samples were examined but correlating dislocation density and stress levels with position relative to microstructure features was possible. It was found that high GND density and stress spots are preferentially accumulated near some grain boundaries, triple junctions and twin boundaries. The correlation coefficients are generally not high (~ -0.2) which we attribute to rather complex and multi-factored interactions involving both elastic and plastic anisotropy which lead to marked internal dislocation substructure within the grains, along with complex and variable grain shapes, micro-texture and subsurface structure which all combine to influence on their final GND density and residual stress distributions. Similar levels of correlation were found at all stages of cyclic deformation examined and also for the single load-unload 'tensile' sample. This may indicate that the GND dislocation structure developed in the first few cycles does not alter much in its general distribution during further cycling.

Dislocation networks tend to be connected spanning across several grains and dislocation bands with high dislocation density bands running along grain boundaries as well as across grain interiors. Such longer range patterning is not obvious and lack quantification in most TEM studies. However, similar to TEM studies, there is also some suggestion that the dislocation structure developed in a grain has some dependence on grain orientation. Orientation dependence of dislocation structure has been reported in the extensive TEM studies by Huang & Winther [56, 57] for monotonic deformation in rolling and tension to larger strains than examined here. Recent HR-EBSD studies of GND structure formed in monotonically deformed copper seem to be in broad agreement and found that $\langle 011 \rangle$ and $\langle 001 \rangle$ oriented grains have more boundary-like dislocation structure and more regular dislocation cell structure is preferentially formed in $\langle 111 \rangle$ oriented grains [45]. Gaudin and Feaugas [58] used TEM to document in detail the orientation dependence of dislocation structures found in 316L stainless steel after tension-tension fatigue.

These GND and residual stress hot spot observations agree well with reported preferential fatigue crack sites observed by optical or scanning electron microscopy as well as predicted mechanistic modelling on various length scales as summarised by recent reviews of fatigue crack nucleation by Sangid [59], Dunne [60] and McDowell and Dunne [61]. Although a detailed fatigue crack mechanism is not yet clear, it is evident that strong correlations have been found between dislocation density and stress hot spots and microstructural features favoured for crack nucleation. In crystal plasticity modelling of cyclic plasticity various so-called fatigue indicator parameters (FIPs) have been suggested as a means of assessing both the likely location and number of cycles required for crack nucleation [61]. These FIPs fall broadly into three types being based either of accumulated slip (plastic strain), or peak tensile stress level (elastic strain), or energy storage (or dissipation). As the test here is controlled by the applied macroscopic stress amplitude and residual stresses are tending to diminish in magnitude it seems unlikely the maximum tensile stress based FIPs will be relevant, unless, as seems unlikely, in some rarer locations not sampled in our EBSD maps higher stress are generated. Either plastic strain or energy storage based FIPs could be feasible. Wan et al [62] have had considerable success rationalising crack nucleation sites and lifetimes in notched fatigue of ferritic steel using an energy accumulation criterion. Local dislocation densities enter their formulation in defining a storage volume based on the mean free path for dislocation slip while the local stress-strain hysteresis allows the work done in the cycle to be evaluated. This is then extrapolated to predict the number of cycles required to accumulate a critical total energy density required for crack nucleation. Thus both stress and dislocation densities are important quantities within the FIP.

5. Conclusions

HR-EBSD was used to qualitatively explore the patterns of dislocation density and intragranular residual stress variations in OFHC polycrystalline copper during the early stages of stress controlled fatigue with a tensile mean stress. These experimental results provide valuable information to modelling community and are an important step to help guide development of fatigue indicator parameters that correctly capture microstructure influence on crack nucleation.

The main findings reported in this paper can be summarised as following:

- The intragranular residual stresses (i.e. variations from the grain averaged mean stress in each grain) was found to decrease in magnitude from 2, to 200, and then again to 2000 cycles. For each sample the stress probabilities were well described by a Gaussian function at small stress magnitudes but with marked deviations at higher stresses.
- The higher stress tails were well-described by $1/\sigma^3$ which is expected for stresses dominated by the closest isolated dislocation. This allowed the total dislocation density to be estimated for each sample. The total dislocation density was highest after 2 cycles (at $\sim 3 \times 10^{15} \text{ m}^{-2}$) but then stabilised at $\sim 7 \times 10^{14} \text{ m}^{-2}$ by 200 and 2000 cycles. The GND densities, calculated over $0.5 \mu\text{m}$ length patches defined by the steps size of the EBSD map, remained fairly constant at $\sim 3 \times 10^{14} \text{ m}^{-2}$ in each of the fatigued samples.

- Quantitative statistical analysis demonstrated that high GND density and high in-plane maximum shear stress tend to be correlated with short Euclidean distances to nearest grain boundaries and triple junctions, and those high GND densities and high stresses tend to be co-located. Similar patterns of correlation were found for each of the fatigued samples and these were also similar to those found in samples deformed in monotonic tension. The correlation coefficients were however only moderate (~ 0.2) indicating that many factors contribute determining the locations of hot spots.
- Dislocation cells and bands were generally observed in the GND dislocation structures. Dislocation cells were formed preferentially in grains with $\langle 111 \rangle$ orientation and dislocation bands exist in grains with $\langle 001 \rangle$ and $\langle 110 \rangle$ orientations. Rather complicated dislocation networks were formed and these often extended over multiple grains indicating that the 'sphere of influence' for any given grain may extend considerably further than just its nearest neighbours.

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Appendix: Estimation of total dislocation by the restricted second moment of probability

Figure A1, schematically illustrating that stress field is dominated by a single edge dislocation toward the patch centre.

Here we summarise evaluation of the total dislocation density, as described in detail by Wilkinson et al .

Imagining that a material can be partitioned into a series of patches with area of A_0 as shown in Figure A1, stress increases significantly toward to the dislocation in the centre. Assuming all dislocations are edge type; the stress near a single edge dislocation with dislocation line along the x_3 axis and Burgers vector magnitude b along the x_1 axis can be expressed as:

$$\sigma_{ij} = \frac{DK_{ij}}{r} \quad \text{Equation 1}$$

Where the r is the radial distant from the dislocation core and θ is the angle from x_1 axis constant D is a constant given in terms of the shear modulus G and Poisson ratio ν by

$$D = \frac{Gb}{2\pi(1-\nu)} \quad \text{Equation 2}$$

The K_{ij} is the angular dependence of the stress. For shear stress σ_{12} , K_{12} is given by

$$K_{12} = \cos(\theta) \cos(\theta) \quad \text{Equation 3}$$

The probability of high stress near dislocation can be established from the fraction area of the patch as shown in Figure 1

$$P = \frac{dA}{A_n} = \frac{rd\theta dr}{A_n} \quad \text{Equation 4}$$

Combining Equation 5 and Equation 6 leads to

$$P(\sigma, \theta) = \frac{D^2 K^2(\theta) d\sigma}{A_o \sigma^3} \quad \text{Equation 5}$$

Integrating around an annulus radius r with width dr allows the angular variation to be averaged, giving the probability as a function of stress:

$$P(\sigma) = \frac{D^2 \langle K^2 \rangle(\theta) d\sigma}{A_o \sigma^3} \quad \text{Equation 6}$$

Where

$$\langle K^2 \rangle = \int_0^{2\pi} K^2(\theta) d\theta \rightarrow \langle K^2 \rangle = \pi/2 \quad \text{Equation 7}$$

From equation 6 we can see that the probability of single dislocation within a patch area A_o is proportional to σ^{-3} . As dislocation density ρ_o is given by $1/A_o$, dislocation density is hence proportional to σ^3 . As the experimental results are rather noisy, here we follow Groma's approach and use the restricted second moment v_2 of probability

$$\begin{aligned} v_2(\sigma) &= \int_{-\sigma}^{+\sigma} P(\sigma) \sigma^2 d\sigma \rightarrow \frac{D^2 \langle K^2 \rangle}{A_o} \ln(\sigma) = D^2 \langle K^2 \rangle \rho_o \ln \frac{\sigma}{\sigma'} \\ &= \frac{(Gb)^2}{8\pi(1-\nu)^2} \rho_o \ln \frac{\sigma}{\sigma'} \end{aligned} \quad \text{Equation 8}$$

Therefore total dislocation density ρ_o can be estimated by determining the gradient of v_2 as a function of $\ln \sigma_{12}$.

Figure 1, EBSD data on annealed copper sample (500μm x500μm). Larger than 10 °misorientation angle between two neighbouring points is defined as a grain boundary. (a) an inverse pole figure map plotted with respect to loading axis [100]. (b) Grain size distribution histogram showing the mean grain diameter is ~10.3μm (averaged by area). (c) an [100] inverse pole figure along loading axis showing weak texture (2x random).

Figure 2, measured stress strain curve for annealed copper samples (a) stress controlled fatigue test showing cyclic stress as a function of cyclic strain on 2 cycles, 200 cycles and 2000 cycles' samples; (b) showing cyclic hardening behaviour by plotting cyclic loop width at mean stress for each cycle as a function of number of fatigue cycles; (c) showing cyclic creep strain as a function of the number of fatigue cycles; (b) SEM micrograph showing fatigue cracks formed on sample free surface after 2000cycles.

Figure 3, Grain morphologies and orientation maps combined with intragranular residual stress and GND density as a function of imposed number of fatigue cycles. (column wise from left): inverse pole figure maps plotted with respect to the loading axis (vertical direction along X_2 axis), grain boundaries marked as black lines were defined as minimum misorientation of 10 degree between two neighbouring points; GND density maps in lines per m^2 . The white areas are unrecovered regions due to low EBSD quality or high misorientation between test points and reference point within that grain. The fourth to sixth columns correspond to three normalised intragranular residual stress component maps ($\sigma_{11}, \sigma_{22}, \sigma_{12}$). Zero residual stress was assigned to the unrecovered white regions as shown in GND density maps.

Figure 4, GND density histograms showing the distribution of GND density in four fatigued samples and 10% monotonically strained sample. 100 bins were used for each dataset to construct this histogram.

Figure 5, the histograms of three intragranular residual stress components: σ_{11} (a), σ_{22} (b) and σ_{12} (c) in annealed, 2 cycles , 200 cycles and 2000 cycles and 10% strained samples. Stresses were normalised by subtracting the mean residual stress within each grain. To highlight tails of distributions curves, the y axes (frequency) are on log scale. Each datasets was binned into 30 bins in stress.

(a)

(b)

Figure 6, (a) probability plot of normal distribution for residual shear stress component σ_{12} . (b) The restricted second restricted moment ($v_2(\sigma)$) as a function of normalised intragranular residual stress σ_{12} on a log 2 scale. Coloured plots are actual data points and the dot lines correspond to fitted gradient to the tails of these curves in order to determine total dislocation density which were calculated and shown as text in this figure.

Figure 7, estimated total dislocation density and GND density on a $\log_{10}$ (dislocation lines per m^2) scale as a function of number of cycles. The map averaged total dislocation density is determined from Figure 6(b). The geometric mean and standard deviation of GND density map are calculated and plot as error bars as indicated by the red line. The error bar of total dislocation density indicates scattering of total dislocation density within individual grains.

Figure 8, data for 2000 cycle sample (similar plots for the other samples are given in the supplementary figures). Maps with colours showing the distance (in μm) from each pixel to the nearest grain boundary (a), or triple junction (d). The remaining plots examine the correlation between position in the microstructure on the vertical axis [represented by distance to nearest grain boundary (b and c) or distance to nearest triple junction (e and f)] and either GND density (b and e) or maximum in plane shear stress (c and f). The grey background intensity shows the number of data points at the corresponding range of distances and GND density or stress. The coloured lines show the median and 5 and 95 percentile binned values of distance with respect to values of GND density or stress along the horizontal axes. Minimum 200 points were required to plot percentiles. The Pearson correlation coefficients were calculated based on all measured points

Figure 9, absolute in plane shear stress as a function of GND density. Dataset was put into 40 bins in both x and y axes. Grey background intensity shows the number of data points in each bin on log2 scale. The coloured lines show the median and 5 and 95 percentile binned values with respect to y axis. Minimum 200 points were required to plot percentiles.

Table(s)

	Cu Annealed	Cu 2 cycles	Cu 200 cycles	Cu 2000 cycles	Cu 10% strained
GND density /Dist to GB	-0.38	-0.36	-0.37	-0.27	-0.29
GND density / Dist to TJ	-0.21	-0.29	-0.38	-0.22	-0.29
Stress /Dist to GB	-0.11	-0.11	-0.2	-0.2	-0.11
Stress /Dist to TJ	-0.08	-0.08	-0.2	-0.2	-0.10
GND density /Stress	0.23	0.24	0.36	0.29	0.30

Table 1, Pearson’s correlation coefficient determined for five samples between GND density, stress and position within microstructures e.g. GBs and TJs. Detailed corresponding distribution figures can be found in supplementary data section.

Supplementary Material

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