Estimation of dislocation density from precession electron diffraction data using the Nye Tensor

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Keywords: Nye tensor, automated crystallographic orientation mapping in transmission electron microscopy (ACOM-TEM), dislocation density, plastic deformation

Abstract

The Nye tensor offers a means to estimate the geometrically necessary dislocation density of a crystalline sample based on measurements of the orientation changes within individual crystal grains. In this paper, the Nye tensor theory is applied to precession electron diffraction automated crystallographic orientation mapping (PED-ACOM) data acquired using a transmission electron microscope (TEM). The resulting dislocation density values are mapped in order to visualize the dislocation structures present in a quantitative manner. These density maps are compared with other related methods of approximating local strain dependencies in dislocation-based microstructural transitions from orientation data. The effect of acquisition parameters on density measurements is examined. By decreasing the step size and spot size during data acquisition, an increasing fraction of the dislocation content becomes accessible. Finally, the method described herein is applied to the measurement of dislocation emission during *in situ* annealing of Cu in TEM in order to demonstrate the utility of the technique for characterizing microstructural dynamics.

1. Introduction

Dislocations in the lattice of crystalline materials alter their mechanical and electrical properties and play a vital role in microstructural evolution [1][2][3][4][5]. Processes of microstructural evolution such as recovery and recrystallization rely on the stored energy from dislocations to drive them [1][2][6]. These relationships can be exploited through controlled thermomechanical processing in order to produce microstructures that optimize desirable properties such as ductility or corrosion resistance. Therefore, the ability to quantitatively measure dislocation densities is crucial to the understanding of deformation, recovery, and recrystallization, and the specific mechanisms by which these processes are driven. The unambiguous quantification of dislocation densities is difficult, however, and current methods based on electron backscatter detection (EBSD) methods can be limited by their spatial resolution and are only applicable to samples that can be analyzed in an SEM. To complement existing techniques and to develop higher resolution methods with higher dislocation density resolutions using automated processes,

we propose a new method to quantify local dislocation densities. This method uses precession electron diffraction (PED) [7][8][9][10] data acquired in a transmission electron microscope (TEM) to generate quantitative dislocation density values based on the Nye tensor dislocation theory [11] for which spatial resolution is limited only by the TEM beam size. It relies on small shifts in orientation rather than relative strains, so that the strain tensor need not be accessible in order to quantify dislocation density.

The prevailing method for automated dislocation density measurements relies on the analysis of EBSD patterns in conjunction with Nye's dislocation density tensor to estimate dislocation densities. Nye's dislocation density tensor [11] relates the dislocation density at a point to the Burgers vector and the dislocation line direction as:

$$\alpha_{ij} = nb_i r_j \quad 1$$

Where $\mathbf{r}$ and $\mathbf{b}$ are the unit and Burgers vectors of the dislocation s , and n is the number of dislocations intersecting with a unit area normal to $\mathbf{r}$. The dislocation density tensor defined this way only represents the net dislocation density for the given volume over which it applies and is related to the curvature of the material at that point. Thus, Nye's dislocation density tensor represents the geometrically necessary dislocations (GNDs) for a given volume or material point. The total dislocation density is a combination of these geometrically necessary dislocations as well as those that don't relate to curvature and thus don't have a net Burgers vector, which are termed statistically stored dislocations (SSDs). Arsenlis and Parks [12] pointed out, however, that the previous statement is only true if the SSD structures are fully contained within the measured volume. As the volume over which a given material is probed is decreased toward the length scale of the dislocation spacing, however, information regarding all dislocations should be accessible [13]. This concept is illustrated in **Error! Reference source not found.** If smaller subdivisions are used, however, the dislocations are seen to contribute locally, even though the alternating curvatures they induce cancel one another out over a larger length scale.

Figure 1. Schematic showing statistically stored dislocation structures inside of a region subdivided into even areas of two sizes (solid and dashed lines). The dislocations shown have opposite Burgers vectors so they will not contribute to lattice curvature from point to point if the large area sections are considered.

As the volume analyzed shrinks to zero, the measured dislocation density tensor approaches the true dislocation density, which is approximated as the L1 norm of the dislocation density tensor normalized by the burgers vector, as shown in equation 2 [14][13]:

Therefore, spatial resolution plays a key role in accessing all of the information regarding dislocation densities, both statistically stored and geometrically necessary.

One drawback to the estimation of dislocation densities using this method is that it ignores curvature that results from residual elastic strain. The development of automated acquisition and analysis of electron backscatter diffraction (EBSD) patterns in scanning electron microscopes (SEMs) [15] has allowed for the application of the Nye tensor model to experimental results [16][17][18], and has allowed for analysis of elastic strain. Kysar, *et al.* [16][17] demonstrated that with a sample in a plane strain condition it is possible to estimate the GND density for individual slip systems as well as the lower-bound GND density using variations of equations 1 and 2 using the point to point changes in orientation determined using EBSD. The cross-correlation technique developed by Wilkinson, *et al.* [19] for the analysis of high resolution EBSD patterns has allowed for the precise quantification of the elastic strain and rotation tensors. Several groups have taken advantage of this newly accessible information to calculate the Nye tensor using Kroner's analysis [13][20][21][22][23][24]. In doing so, however, Wilkinson, *et al.* determined that the contribution of elastic strain is insignificant compared to lattice rotation in contributing to overall lattice curvature of plastically deformed metals [20], justifying the application of Nye's original treatment without the addition of terms to incorporate elastic strain [21][22]. Likewise, recent work by Ruggles and Fullwood shows that for data acquired using EBSD incorporation of elastic strain measurements in addition to rotation does not significantly increase the accuracy of the calculation [13].

Other techniques exist for mapping elastic and plastic strain using EBSD patterns, including image quality analysis [25][26] and local misorientation mapping methods such as kernel average misorientation (KAM) analysis, which was introduced by D.P. Field, *et al.* [27][26]. While these methods are less quantitative than the cross-correlation approach with respect to dislocation density measurement, they are able to yield data that can be used to estimate the relative dislocation densities. Another method for quantifying dislocation densities from EBSD is the weighted Burgers vector analysis developed by Wheeler, *et al.* [28]. The weighted Burgers vector is defined as the sum of the densities of each type of dislocation multiplied by their Burgers vectors. This method has the advantage that there is no information about the Z axis required to define it fully.

Despite recent improvements in resolution, conventional EBSD on bulk samples in SEMs is limited to a minimum spatial resolution of approximately 20-40 nm depending on the material being examined [29][22][30]. This drawback, along with its nature as a surface characterization technique, makes SEM less than ideal for measuring dislocations. TEM is an obvious choice as a replacement technique due to its superior resolution and because, being a transmission technique, it is capable providing images of the dislocation structure to accompany the quantitative evaluation using the Nye tensor. This allows for a visual comparison of the resulting GND

density estimate with the dislocations present in the accompanying TEM micrograph and provides useful input for scanning parameters, such as approximate dislocation spacing and subgrain sizes. This point was made by Wilkinson, *et al.* [19], who touched on the possible advantages of using diffraction patterns acquired in TEM when the cross-correlation EBSD technique was first described. The one drawback of using a transmission technique is that the sample has two free surfaces so more relaxation of the defect structure can occur. It is important to note that advancements in transmission Kikuchi diffraction (TKD) have shown promise for decreasing the spatial resolution of EBSD systems below 10 nm for electron-transparent thin foils, making TKD another viable alternative [30]. Since the specimens for TKD must be electron-transparent the sample preparation for this technique is similar to that of TEM. It is critical to note, however, that while TKD provides advantages in angular resolution, it still lacks the ability to directly image the internal defect structures simultaneously.

One approach to dislocation density measurement in TEM is the analysis of precession electron diffraction (PED) patterns [7]. PED is diffraction acquired using a hollow cone of illumination produced by rapidly precessing the beam in a circle [7]. The use of PED allows for quasi-kinematical diffraction conditions that make visible the reflections from the higher order Laue zones (HOLZ) that contain information about the structure parallel to the optic axis that is typically inaccessible in spot diffraction patterns [31].

Automated crystallographic orientation mapping in TEM (ACOM-TEM) [8][9][10] provides a potential source of input data for dislocation density calculations based on the Nye tensor; when used in conjunction with PED, it provides three-dimensional orientation information as well as the displacement in two dimensions. In this paper, we apply existing methods for quantifying plastic deformation from orientation data and present a method for estimating and mapping the dislocation density in TEM specimens using the Nye tensor utilizing Euler angle and displacement input from ACOM-TEM [32]. By calculating the best fit of the tensor describing the contortion of the lattice for each point with respect to its nearest neighbors, changes in the dislocation density from point to point can be measured on a nanometer length-scale. A similar methodology was first described by Pantleon [18] using conventional EBSD data as inputs. An alternate approach to applying the Nye tensor to ACOM-TEM to quantify dislocation density was recently demonstrated by Ghamarian, *et al.* [33]. That method, which was developed concurrently with the one described herein, is similar in approach but the dislocation density maps produced appear very different. The primary difference is the use of Kuwahara filtering to manipulate the orientation data prior to calculating the Nye tensor, which is addressed in Section 4.4. Additionally, it is unclear from [33] how the Nye tensor values for each individual point are averaged and assigned. This analysis technique is capable of accessing both GNDs and SSDs due to the scale of the applied probe. It has been applied to the study of the effect of dislocation densities on microevolution processes such as grain rotation, boundary migration, and other microstructural transitions [34]. Maps produced using this technique are consistent with TEM images of dislocation structures taken in the analysis regions, and the dislocation density values

calculated are on the same orders of magnitude with those from literature [35][22][36]. The results generated by this method presented herein for quantification of both geometrically necessary and statistically stored dislocation content is compared to those generated by related methods for analyzing dislocation dependent microstructural events, such as kernel average misorientation and local orientation spread.

2. Material and Methods

2.1 Sample Preparation

Cold-rolled microcrystalline oxygen-free electronic copper (99.99% pure) half-inch plate (McMaster Carr) was cut into 1.25 x 1.25 cm strips and annealed for one hour at 540°C to achieve a fully recrystallized microstructure. The strips were then subjected to various additional rolling reductions ranging from 5 to 50 percent in order to achieve a range of dislocation densities. Some specimens were subjected to partial annealing at temperatures ranging from 400-720 °C in order to produce microstructures in the various stages of recovery and recrystallization. TEM disc specimens were cut from the material perpendicular to the rolling direction and prepared using conventional metallographic techniques and jet polished using a Struers TenuPol-5 electropolisher. In addition, site-specific lift-outs of individual grain boundaries were prepared using an FEI Strata DB235 focused ion beam (FIB).

2.2 Precession Diffraction Pattern Acquisition

The NanoMEGAS SPINNING STAR™ precession electron diffraction and ASTAR™ ACOM-TEM systems, integrated into a JEOL 2100 Lab6 TEM, were used to acquire the orientation data used as the input for GND density calculations [32]. A precession angle of 0.60° was utilized for all experiments. Step sizes of 10.40 nm and 3.90 nm for both x and y directions were used. The beam spot size used was 10 nm. The patterns were acquired and indexed automatically following a method first described by Rauch and Veron [8].

Because the step size and probe size for this setup are not linked there is a potential for error due to under- or over-sampling depending on the acquisition parameters. Likewise, the acquisition parameters ought to have a direct effect on the fraction of the dislocation that is geometrically significant at the length-scale being considered as previously discussed. A sampling model was produced to evaluate how these variables affect the values produced. As shown in Figure 2A, the model takes measurements of the average orientation across a width D that are a distance Δ apart. The specimen being measured contains an array of equally spaced dislocations d apart that have either positive or negative signs assigned randomly. For a complete description of the

sampling model see Appendix A. As can be seen in Figure 2B, the fraction of dislocations detected (ρ/ρ_0) increases as Δ is decreased. For all spot sizes tested the relationship between ρ/ρ_0 and Δ is the same until Δ/d approaches one. For spot sizes approaching or decreasing beyond d this trend continues as ρ/ρ_0 approaches unity and levels off, meaning the total dislocation content is measured without over-sampling. No combination of Δ and D lead to an overestimation of dislocation density and the systematic error caused by under-sampling can be corrected for, especially with iterative measurements with varied acquisition parameters.

Figure 2. A) Schematic of sampling model. B) Fraction of dislocation density measured by simulation versus step size for various spot sizes. Each line is the average of 10 iterations with randomized dislocation signs.

2.3 Local Orientation Spread and Dislocation Density Analysis

The orientation data was analyzed using the TSL OIM Analysis software package developed by EDAX. Using this software, maps of local orientation spread and kernel average misorientation, metrics typically used to assess local deformation in SEM specimens analyzed using EBSD, were generated in order to qualitatively visualize internal strains due to lattice curvature. This serves as a rough approximation of GND density because any curvature within an individual grain must be accommodated by dislocations [26]. Local spread is calculated by treating each point in the scan as the center of a kernel of n nearest neighbors then averaging the misorientation between each pair of points within the kernel and assigning the mean value to the center point. Kernel average misorientation is calculated similarly, but instead of considering each pair of points, only the misorientation between the center point and the other points are used. A kernel size of $n = 3$ and exclusion angle of 5° was chosen for all data for the purpose of consistency. Schematic representations of these calculations are shown in Figure 3 along with an example map for each. Although it has not previously been performed with crystal orientation data acquired in the TEM, this method has been used extensively for the analysis of EBSD data as discussed in Section 1 [27][26].

Figure 3. A&B) Schematic representations of kernel average misorientation and local spread calculations, respectively. C&D) Kernel average misorientation and local orientation spread maps of a region adjacent to a $\Sigma 3$ grain boundary in Cu.

In order to obtain a more accurate, quantitative estimate of the GND density beyond local orientation changes, the contortion tensors relating each point to its eight nearest neighbors were calculated. Each point was considered as the center of a kernel with a radius of one and the tensor components relating the eight neighbors in the kernel to it were determined. A least-squares fitting was then applied to calculate a single contortion tensor that best describes the relationship between the center point and its neighbors. These least-squares contortion values were then used to calculate a single Nye tensor that describes the state of dislocation at each point. While there exists a wide range of approaches to calculating a set of Nye tensor values to

describe a given unit volume because each set of tensor values describes the dislocation content required to accommodate the disorientation between two known points, the method described herein avoids both directionality (i.e. comparing each point to its right-hand neighbor) and the use of a reference or average orientation. It is not clear that previous efforts [33] addressed this critical point. Once the Nye tensor describing each point has been calculated, equation 2 is used to estimate the lower-bound GND density based on the assumption that all dislocations have a $1/2\langle 110 \rangle$ Burgers vector [22][13] since Cu is FCC. For other crystal structures where only a single family of Burgers vectors is present (such as $1/2\langle 111 \rangle$ in BCC), this assumption is easily adapted. For crystal structures with more than one family of dislocations, such as HCP, a weighted average Burgers vector must be calculated [13]. Only five of the nine tensor components were directly accessible from the data as discussed in detail in Section 3 so only those components were taken into consideration. Contour plots of the estimated lower-bound GND density were generated in OriginTM for direct comparison with TEM micrographs of the regions analyzed.

3. Nye Tensor Calculation

The Nye tensor defined in equation 1 can also be described in terms of the contortion K , as follows:

$$\alpha_{ij} = K_{ji} - \delta_{ij}K_{kk} \quad 5$$

Where K_{kk} is in the Einstein summation notation implying that $K_{kk} = \sum_i K_{ii}$. $\delta_{ij} = 1$ for $i = j$ and $\delta_{ij} = 0$ for $i \neq j$. Nye defined the contortion as a second-rank tensor:

$$K_{ij} = \frac{\partial \varphi_i}{\partial x_j} \approx \frac{\Delta \varphi_i}{\Delta x_j} \quad 6$$

$\partial \varphi_i$ is the three-dimensional rotation vector and ∂x_j is the displacement vector. These are generated using ACOM-TEM. First, the axis-angle pair for the disorientation between each point and its neighbors is calculated. The rotation vector $\partial \varphi_i$ is then approximated as the product of the axis and angle according to infinitesimal rotation theory [37]. ∂x_j is the displacement between the steps being compared. Equation 6 is undefined whenever $j = 3$ because the displacement along the z-axis is zero. These undefined values for K_{i3} will carry through when K is plugged into equation 5, which is written explicitly as:

$$\alpha_{ij} = \begin{pmatrix} -K_{22} - K_{33} & K_{21} & K_{31} \\ K_{12} & -K_{11} - K_{33} & K_{32} \\ K_{13} & K_{23} & -K_{11} - K_{22} \end{pmatrix}. \quad 7$$

From this expression it is clear that the components α_{11} , α_{22} , α_{31} , and α_{32} will be undefined with the present inputs (components shown in gray).

4. Results and Discussion

4.1 Local Orientation Spread

Maps of local orientation spread (LOS) were produced as a preliminary method for qualitatively assessing strain using ACOM-TEM data [32] and to compare with studies performed by Field *et al.* [38][26]. This technique provides a rough estimate of the strain in the lattice due to local changes in the crystal orientation caused by the presence of dislocations or long-range elastic strain gradients. A LOS map is shown in Figure 4 along with a corresponding TEM micrograph and orientation map. The LOS maps are normalized to the minimum and maximum orientation spread values present within the dataset, in which high angle grain boundaries (HAGBs) are shown in black, $\Sigma 3$ boundaries in red, and low angle grain boundaries (LAGBs, 2-15°) in teal.

At high magnification, the width of the boundary is able to be resolved, and thus the orientation is viewed clearly as a gradient across a finite distance (as opposed to an abrupt jump from point to point in a scan line). As a result, the TSL software fails to identify the 7° LAGB indicated with the solid arrow in Figure 4 as a grain boundary, instead showing it as a wide orientation

274 gradient. This can be seen as a second peak in the LOS distribution (

275

276 **Figure 4D**), which typically follows a log-normal distribution when LAGBs are not present, or at
277 spatial resolutions where their width is not resolved like in EBSD. It is important to note that
278 while some of the strain concentrations highlighted in the LOS map (Figure 4C), such as the one
279 indicated with the dotted arrow are readily visible in the corresponding TEM micrograph (Figure
280 4A), others, such as the feature indicated with the dashed arrow, are not resolved. In addition, the
281 effects of bend contours are not present in the LOS maps, thus reducing the level of noise in the
282 image and facilitating dislocation analysis. The ability of this analysis to reveal dislocation
283 structures more readily than by TEM imaging alone demonstrates the importance of developing
284 characterization techniques capable of measuring dislocation content that can be utilized in a
285 complimentary fashion with TEM.

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288

289 **Figure 4.** A) TEM micrograph of triple junction and LAGB. B) Corresponding inverse pole
290 figure map. C) Local orientation spread ($3n$). D) Number fraction distribution of local orientation
291 spread.

292

293 4.2 Nye Tensors and GND Densities

Lower bound GND density (LB-GNDD) maps generated by the method described herein yield a quantitative view of dislocation structures. They show advantages over LOS maps in that the values correspond directly to the defects present in the specimen whereas the local spread only provides a relative comparison. Additionally, not all features of the dislocation structure are resolved by the LOS maps. **Figure 5** shows a partially recovered region near a HAGB with corresponding LOS and LB-GNDD maps. The two maps are qualitatively similar, with small differences arising from the different number of nearest neighbors and maximum angles included in the calculations, but the GND density map provides a better quantitative representation of the defect distribution.

The GND density is plotted on a log scale in order to show the full range of values, which are high compared to those reported in the literature from EBSD data [35][22] (10^{13} - 10^{16} m⁻²). This discrepancy can be attributed to the very fine step size used relative to EBSD measurements as well as differences in probe size and angular resolution [22], as is addressed in detail in the following section. Another factor that could contribute to the high density values is that all of the regions being examined are adjacent to grain boundaries. GND mapping in EBSD typically shows the highest densities in the regions along the boundaries, with low densities in the grain interiors. Another factor that contributes to the high GND density values reported herein is the minimum angular resolution of the indexing software used to determine the orientation at each point. The minimum resolution for the current indexing method with the current input parameters is approximately 0.5°. This is determined by the angular spacing between template patterns used for indexing. Any rotation that is smaller than this value but still detected by the software will be inflated to this minimum, artificially increasing the densities reported. Improvements to the angular resolution of pattern indexing will reduce the significance of this effect. A method was described recently to use the intensity of the spots to interpolate between template orientations in order to optimize the index quality [39]. By applying this orientation refinement algorithm, a resolution better than 0.3° was reported. Application of this method in conjunction with the present analysis method is discussed in Section 4.4. Nonetheless, angular resolution is currently the primary drawback of using PED-ACOM data for strain mapping [33] and the main advantage of TKD as an orientation mapping technique for thin specimens [30]. One potential path forward for dislocation mapping in TEM at high angular resolution is the use of automated CBED [40].

The step size used for the acquisition of orientation data is more than an order of magnitude beneath the minimum dislocation cell size for copper of ~250 nm as observed in literature[41][42]. As a result, this technique is able to resolve the walls and interiors of dislocation cells, even in metals that have undergone severe plastic deformation, as shown in **Figure 5C&E**. It should be noted that the step sizes used in the analysis shown in **Figure 5C** were on the same order of magnitude as the dislocation spacing. As such, the measured volumes include fractions of defect structures that, taken together, would be considered as SSDs. As discussed in detail in Section I, if the complete SSD structure is not contained within the volume used for the calculation of the Nye tensor, then those dislocations will be accessible using this

method. As a result, although the values reported herein are referred to as GNDs, they ought to represent a larger fraction of the total dislocation density than GND densities calculated using larger probe volumes. In **Figure 5C** the dislocation cell walls are clearly outlined, as expected, but variations in dislocation density within the interiors of the individual cells due to SSDs are also resolved as can be seen clearly in the blown up region of the map shown in **Figure 5E**.

It is important to note that if a high GND density is calculated based on the misorientation where it is clear from microscopy that none is present then the curvature in the lattice at that point must be due to elastic strain.

It is not always clear, however, whether dislocations are present in a given region due to the effect of diffracting conditions in the TEM on the visibility of defect structures. Elastic strain cannot be measured using the current approach, although it can be extracted from an on-axis nanobeam diffraction pattern by cross-correlation with a reference pattern from an unstrained region. If significant elastic strains are present they will also contribute to the inflation of GND density values for the present methodology since they are not taken into consideration in the calculation so all orientation changes are attributed to dislocations.

The primary drawback of the present analysis is the unknown thickness of the specimen along the z-axis, which is similar to the difficulties encountered with EBSD being a surface technique. This results in only five of the components of the Nye tensor being directly accessible [18].

Figure 5. A) TEM micrograph of a HAGB in a region that has undergone a partial recovery process. B) Local orientation spread map produced for the same region. C) Estimated lower-bound GND density map. D) Expanded view of the region outlined in A. E) Expanded view of region outlined in C, which corresponds to D. Dashed arrow indicates LAGB. Solid arrows indicate dislocation cell walls.

In order to assess the effect of step size used for data acquisition on the maps produced by this method, individual regions were scanned at a range of step sizes. Decreasing the step size independent of the probe size was shown to improve the spatial resolution of the technique. Most importantly, as predicted by the sampling model, the fraction of dislocations measured increases as Δ/d is decreased. Jiang, *et al.* [22] observed the same phenomenon for data acquired using EBSD, noting that the density values calculated decrease when the data is binned to a lower spatial resolution.

An initial step size of 10.4 nm was chosen because it matches well with the minimum effective probe size of the TEM being used. Because the defect spacing and feature size is on the same order of magnitude, however, a smaller step size of 3.9 nm was used to analyze the effect of oversampling. As can be seen in Figure 6E&F, decreasing the step size results in an increased spatial resolution, as expected. The resulting maps reveal the GND structure in greater detail, removing much of the noise present in the maps produced with the larger step size. These same effects are clear from the LOS maps shown in Figure 6C&D, where the qualitative details of the defect structure are much more visible at the smaller step size. The normalized dislocation density probability distribution for a given area remains approximately the same regardless of step size (Figure 6G and H), following a log-normal relationship that is nearly identical with dislocation density probability distributions reported in the literature from EBSD-based analysis [43]. This indicates that a calibration factor can be developed in order to produce accurate, repeatable measurements of the dislocation density distribution regardless of the step size used. The average density for the first 780 by 780 nm area of each scan is $3.62 \times 10^{15} \text{ m}^{-2}$ for the 10.4 nm step and $7.34 \times 10^{15} \text{ m}^{-2}$ for the 3.9 nm step. These values were calculated by averaging the point densities across this area. This increase in measured dislocation density is due to the fact that an increasing fraction of dislocations are geometrically significant as the length-scale of the measurement is decreased due to short-range fluctuations in orientation produced even by dislocations traditionally considered to be SSDs as illustrated in Figure 1. This effect is especially drastic as step sizes decrease beyond the average dislocation spacing. A dislocation density of $7.34 \times 10^{15} \text{ m}^{-2}$, the value produced using the 3.9 nm step, is equivalent to a mean dislocation spacing of 11.7 nm, so the increase in measured dislocation density when the step size is decreased from 10.4 to 3.9 nm is expected according to the results of the sampling model shown in Figure 2B. The step size for acquisition can be further reduced, potentially down to a fraction of a nanometer. Doing so without also reducing the probe size of the TEM will result in further oversampling, with each pattern containing much the same information as the patterns acquired adjacent to it. Since this method relies on orientation measurements rather than a direct count, oversampling does not lead to over-counting, assuming that the orientation assigned is a weighted average of the distribution of orientations contained within the probe volume. If the measurement spacing is further reduced, eventually a critical step size (Δ_c) will be reached where all dislocations are accessible. This Δ_c is not a constant as it depends both on the minimum dislocation spacing present in the specimen and the probe size being used. Oversampling will, however, lead to increased density maxima and a higher number of points with low density

400 values since the nominal area over which the density is calculated decreases but the probe area
401 stays the same. Since dislocations tend to be concentrated into tangles and cell walls, density
402 values from measurements taken in such regions will increase as step size is decreased because
403 the same number of dislocations will be probed over a smaller nominal area. The interiors of
404 dislocation cells will, however, contribute a higher fraction of data points as step size is
405 decreased. The interaction of these two effects is such that the average dislocation density for a
406 region should remain constant except for the effects of accessing SSDs as previously discussed.
407 One should note for experimental planning that decreasing the step increases acquisition time
408 scans.

Figure 6. A&B) TEM micrographs of the scan area, where B is at an increased magnification. C&D) Local orientation spread maps corresponding to the region shown in A & B with step sizes of 10.4 and 3.9 nm, respectively. E&F) Maps of estimated lower-bound GND density produced from the same data as C & D. G&H) Normalized dislocation density probability distributions and cumulative distributions, respectively. Area 1 is the region shown in A-F.

4.4 Noise Effects and Image Enhancement

Some amount of noise is present in the orientation data due to the angular resolution limit of the indexing technique. This is of particular concern when the acquisition step size is varied since

the minimum resolved dislocation density is inversely proportional to the step size [22]. In order to determine what role, if any, noise due to angular resolution plays in the high dislocation density values calculated from data acquired at very fine step sizes, three approaches were taken.

First, various amounts of noise were added to an orientation data set prior to Nye tensor analysis. Each data point had a 50% probability of being rotated a set amount – analogous to a minimum angular resolution – about a random axis of rotation. As can be seen in Figure 7, increasing the amount of noise in the orientation data causes an increase in calculated dislocation density. Unlike the increased densities calculated at reduced step sizes, there is a significant change in the cumulative probability function associated with this shift. Additionally, whereas with decreased step sizes the dislocation structures become sharper and more clearly correspond with the features visible in the micrograph, increasing the noise causes the features to be obscured as one would expect. Furthermore, it takes an addition of 5° random rotations to produce an increase in dislocation density of approximately $1 \times 10^{16} \text{ m}^{-2}$, approaching the scale of the densities calculated at the smallest step sizes used.

Figure 7. A) Cumulative probability distribution functions for the dislocation density data shown in C) with various levels of noise added. B) inset region from A showing the crossover at $\rho/\rho_{\text{avg}} = 1.5$. The higher the level of noise, the less likely that a given point will have a density below this

value. C) GND density map produced from raw data (no noise added). D) GND density map produced from orientation data with a 0.5° rotation added. E) GND density map produced from orientation data with a 5° rotation added.

Second, a low-pass Gaussian discrete Fourier filter was applied to dislocation density data sets in order to quantify the effects of noise on the values and to improve the image quality. As can be seen in Figure 8C&D, this filtering reduces the visual noise in the image by removing any high frequency shifts in dislocation density such as those associated with the angular resolution limit. Quantitatively, it produces a decrease in mean dislocation density on the order of 10^{11} m^{-2} , even for scans taken at reduced step sizes. This indicates that high frequency noise is not responsible for the increase in calculated densities in this data relative to data acquired at larger step sizes.

Finally, two methods were used in an effort to enhance the effective angular resolution of the technique. First, a Kuwahara filter was applied to the orientation data after indexing was completed according to the method described by Humphreys, *et al.* [44]. A kernel size of five pixels was used for filtering, with the quadrant with the lowest variance being assigned as the new orientation for the center point in the kernel. This type of filtering minimizes small fluctuations in the data such as those caused by high frequency noise while sharpening edge features. It was chosen because recent work by Ghamarian, *et al.* suggests that this is an ideal way to enhance the angular resolution of ACOM data. By comparing Figure 8E&F with A&B, however, it is apparent that in this case Kuwahara filtering resulted in substantive changes to the calculated dislocation structure. Moreover, due to the counterbalancing effects of smoothing small fluctuations and emphasizing large ones, the filter does not cause a systematic decrease in the average dislocation density and in some cases actually causes the average to increase.

The second method used to enhance the angular resolution of the ACOM data was an interpolation algorithm described in detail recently by Rauch and Veron [39]. By interpolating between the positions and intensities of spots in the neighboring template patterns that best fit the experimental diffraction pattern, the orientation can be estimated with more precision (approximately 0.3°) than the sampling of orientation space represented by the template bank. As can be seen in Figure 8G&H, the morphology of the calculated dislocation structures did not change significantly when this refinement was utilized. Most importantly, this method does not cause a systematic decrease in the average dislocation density. This indicates that the increases in dislocation density seen at small step sizes is not due to angular resolution constraints because if that were so then improving the angular resolution while holding the step size constant ought to cause a corresponding decrease in density values.

Figure 8. A&B) GND density maps produced from raw data (no filtering). C&D) GND density maps for the same regions as A&B produced from data that has been filtered using discrete Fourier transform analysis. E&F) GND density maps produced using Kuwahara filtering. G&H) GND density maps produced using interpolation method to refine the angular resolution.

4.5 Dislocation Motion and Dislocation-Driven Processes

By applying this technique before and after an external stimulus during *in situ* processing, such as the application of heat or strain, motion (e.g, annihilation or emission) of dislocations can be quantified. One example of this, shown below, is the quantification of grain rotation through the emission of dislocations from a triple junction during annealing. Figure 9 shows a region adjacent to a triple junction in Cu before, during, and after annealing *in situ* at 720 °C. As shown in the video stills in Figure 9D, during the annealing process dislocations were emitted from the triple junction which then glide approximately 800 nm into the matrix of the lower-right grain before piling up. As a result of this process the corner of the grain is rotated, creating a sub-grain with a disorientation of 3.5° relative to the original matrix grain as shown in the misorientation vs. displacement plot (Figure 9E). As can be seen in Figure 9C, the dislocation density map from prior to this process indicates that the grain had a uniform dislocation distribution. The data recorded after heating, on the other hand, clearly shows the build-up of dislocations where the sub-boundary has formed (Figure 9G&H). Grain rotation is just one example of a process of microstructural evolution that can be informed by this technique. The mechanism and driving force for twinning is of particular interest and is the subject of a parallel work using this methodology [34].

Figure 9. A&F) TEM micrographs showing two triple junctions in a Cu foil before and after *in situ* annealing, respectively. B&G) LOS maps corresponding to the images shown in A and F, respectively. C&H) Dislocation density maps corresponding to the images shown in A and F, respectively. D) Video stills from *in situ* heating showing dislocations being emitted and piling up in the region outlined in A and F. E) Plot of misorientation with respect to origin along the lines shown in A and F. I) TEM micrograph showing the area outlined in E at higher magnification. J) Region of GND map shown in H corresponding to the area shown in I.

5. Conclusions

The primary goal of this work was to demonstrate the viability and utility of applying local misorientation analysis methods and Nye's dislocation theory to the data acquired by precession-enhanced ACOM-TEM. Two methodologies for quantitatively mapping the state of deformation

using ACOM-TEM data have been demonstrated – local orientation spread mapping and Nye LB-GNDD mapping.

The local orientation spread approach provides relative values of strain from point to point for a given region. It is a fast, convenient method for discerning relative dislocation density values from PED-ACOM data that can be carried out using commercially available software. The values produced are essentially a scalar estimation of local strain that can be compared directly from sample to sample for a given material and consistent acquisition parameters. This method is applicable to all known crystal structures as long as their diffraction patterns can be indexed accurately. The primary drawback of this method is that the contributions of elastic and plastic strain cannot be decoupled from one another so the individual strain tensor components cannot be determined.

The estimated lower-bound GND density approach provides an approximate measurement of the dislocation density at a given point. These values are directly comparable to one another because they are a measurement of the number of defects present. The method described herein is similar in approach to the technique described recently by Ghamarian, *et al.* [33] and produces calculated defect densities on the same orders of magnitude. The application of a hard angular limit for inclusion in calculations and least squares fitting of neighbor points in the present work instead of Kuwahara filtering leads to a very different final product. This method is easily adapted to BCC materials and other structures that contain only a single family of dislocations. It can be adapted to other crystal structures through the calculation of a solution to the Nye tensor that minimizes the line energy of the dislocations while accommodating the curvature [45] that describes all the dislocation types present and their relative frequency. Due to its quantitative nature, the analysis described herein is a powerful tool for measuring dislocation densities in TEM specimens that has the potential to inform many different processes of microstructural change, especially when combined with *in situ* TEM studies. Moreover, this method is not limited to PED-ACOM data; without modification it can be applied to EBSD data acquired in the SEM.

Due to the short length-scale of the measurements described herein, the dislocation densities reported ought to take into account some fraction of the SSDs present as well as the GNDs. Measurements of the same regions at multiple step sizes indicate that this is the case. A simplified sampling model was produced in order to approximate the effects of acquisition parameters on the resulting measurements. This model demonstrated the potential of this type of analysis to access the complete dislocation content of a specimen when a suitable combination of spot and step sizes are used in acquisition. A more detailed mathematical model must be developed in order to estimate more precisely what fraction of the total dislocation density is being measured using a given set of acquisition parameters.

Increased accuracy for both methods described herein can be achieved by the improvement of the indexing for the diffraction patterns in order to increase angular resolution. Likewise,

improved spatial resolution can be achieved using a finer probe and step size. The effects of elastic strain present in the material on the calculated dislocation densities must be determined in order to ensure the accuracy of the calculated densities, although previous work has found their contribution to be negligible for similar methods applied to EBSD. This could be achieved by the analysis of diffraction spot positions relative to a reference or simulated pattern of the same orientation, or by utilizing automated CBED, in which the strains are more readily accessible.

Coupling this method of quantitative dislocation analysis with orientation-based methods for qualitatively resolving strain local strain yields a superior understanding of microstructural events and thus holds key advantages for spatially resolving dislocation- dependent phenomena in various materials.

Acknowledgements

Authors ACL and MLT gratefully acknowledge funding from the United States Department of Energy Basic Energy Sciences (DOE/BES) under the Early Career program through contract DE-SC0008274, funding from the National Science Foundation's Faculty Early Career Program under contract #1150807, and funding from the Department of Energy's Nuclear Energy University Program under contract NE0000315.

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Appendix A – One-dimensional model of orientation-based sampling for dislocation measurement

In order to examine the effects of the data acquisition parameters on the measured dislocation density using orientation-based sampling, a simple model was developed using MATLAB®. The sampling model begins by defining a one-dimensional specimen of length L with dislocations evenly spaced a distance of 1 arbitrary unit apart, starting at a user-defined offset from the edge of the specimen. In this way, the dislocation density is set equal to 1. Each dislocation is assigned either a positive or negative sign, which constitutes its Burgers vector. Two adjacent dislocations with opposite signs constitute a statistically stored dislocation dipole, whereas consecutive dislocations of the same sign are geometrically necessary. Various schemes for assigning signs to the dislocations were tested such as all positive (all GNDs), alternating signs (all SSDs), every third dislocation negative, and random assignment. Each dislocation causes an arbitrary rotation of equal magnitude to the specimen with the direction of the rotation dependent on the sign. Once the simulated structure has been generated, N measurements of the average orientation of the specimen across a probe diameter D are taken at a distance Δ apart. D is equivalent to the beam spot size and Δ is equivalent to the step size for diffraction pattern acquisition. A simplified contortion tensor K was then calculated as the difference in orientation between the two nearest neighbors of each measurement divided by the distance between those two measurements (2Δ). The measured dislocation density for the specimen was calculated according to:

$$\rho_{tot} = \sum |K| * \Delta / L \quad (A1)$$

In addition, the fraction of the measured density that is attributable to GNDs was calculated according to:

$$\rho_{GND} = \sum K * \Delta / L \quad (A2)$$

By comparing the values of ρ_{tot} and ρ_{GND} the combinations it was shown that dislocations that are not geometrically necessary were resolved, especially at spot sizes lower than the dislocation spacing.