Quantifying the Influence of Twin Boundaries on the Deformation of Nanocrystalline Copper using Atomistic Simulations

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Abstract

Over the past decade, numerous efforts have sought to understand the influence of twin boundaries on the behavior of polycrystalline materials. Early results suggested that twin boundaries within nanocrystalline face-centered cubic metals have a considerable effect on material behavior by altering the activated deformation mechanisms. In this work, we employ molecular dynamics simulations to elucidate the role of twin boundaries on the deformation of $\langle 100 \rangle$ columnar nanocrystalline copper at room temperature under uniaxial strain. We leverage non-local kinematic metrics, formulated from continuum mechanics theory, to compute atomically-resolved rotational and strain fields during plastic deformation. These results are then utilized to compute the distribution of various nanoscale mechanisms during straining, and quantitatively resolve their contribution to the total strain accommodation within the microstructure, highlighting the fundamental role of twin boundaries. Our results show that nanoscale twins influence nanocrystalline copper by altering the cooperation of fundamental deformation mechanisms and their contributed role in strain accommodation, and we present new methods for extracting useful information from atomistic simulations. The simulation results suggest a tension-compression asymmetry in the distribution of deformation mechanisms and strain accommodation by either dislocations or twin boundary mechanisms. In highly twinned microstructures, twin boundary migration can become a significant deformation mode, in comparison to lattice dislocation plasticity in non-twinned columnar microstructures, especially during compression.

Keywords: Molecular Dynamics, Twinning, Dislocations, Grain Boundaries, Polycrystalline Material

1. Introduction

Improving the mechanical properties (e.g., strength, ductility, fatigue resistance, etc.) of materials for advanced functionality has motivated recent work to investigate the influence of altering a material's substructure. One common approach for improving strength in metals is to reduce the average grain size, as described by the Hall-Petch relationship (Hall (1951); Petch (1953)). The Hall-Petch effect (i.e., strengthening with decreasing average grain size) in polycrystalline materials is based on the idea that interfaces between grains (i.e., grain boundaries (GBs)), serve to block the migration of lattice dislocations and suppress conventional dislocation strain accommodation. Higher resolved stresses are then required to overcome these obstacles, thereby, increasing the strength of a material. However, early findings showed that as material strength increased due to grain size reduction, there is a simultaneous decrease in material ductility (Gleiter (1989); Koch (2003); Ma (2003)). Therefore, recent efforts have focused on simultaneously improving strength and ductility in metallic materials using a variety of approaches (Ertorer et al. (2009); Karimpoor et al. (2003); Ma (2006); Shen et al. (2008); Valiev et al. (2002); Youssef et al. (2005); Zhao et al. (2008)).

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As grain size is reduced to create nanocrystalline (NC) materials, the influence of GBs on macroscopic behavior is more substantial and alternative deformation mechanisms begin to operate (Capolungo et al. (2007); Chen et al. (2003); Derlet et al. (2006); Frøseth et al. (2004, 2005); Kumar et al. (2003); Liao et al. (2003); Meyers et al. (2006); Schiotz (2004); Schiotz and Jacobsen (2003); Shan et al. (2004); Tschopp and McDowell (2008); Van Swygenhoven and Derlet (2001); Van Swygenhoven and Weertman (2006); Wang et al. (2012); Yamakov et al. (2004); Zhang et al. (2013); Zhu and Lu (2012)). However, as Schiotz and Jacobsen (2003) noted, a transition from dislocation-dominated to GB-mediated plasticity in Cu occurs at an average grain size that exhibits maximum strength in NC systems. These results indicate that there are inherent length-scales (i.e., average grain size and mean dislocation slip distance) that affect deformation and the role of nanoscale interfaces in NC metals is fundamental to material behavior. Nanoscale interfaces, such as coherent twin boundaries (TBs), have been the subject of recent endeavors aimed at improving strength without limiting other physical properties (Karimpoor et al. (2003); Lu et al. (2009); Ma et al. (2004); Wei (2011); Zhao et al. (2006); Zhu et al. (2011)). This is in part motivated by work from Lu et al. (2004) showing that nanoscale growth twins in NC Cu led to significant improvement in strength while preserving electrical conductivity. These findings suggest that coherent TBs within nanoscale grains hinder the migration of lattice dislocations and lead to strain hardening, similar to other GBs, but do not decrease electrical conductivity. Furthermore, recent findings by Wang et al. (2007) have shown that dislocations can preferentially be emitted from the intersection of a GB and TB to induce TB migration in Cu during tensile deformation. This indicates the potential role of nanoscale twins to affect the deformation behavior in NC metals during plasticity by altering the barrier for dislocation nucleation and serving as an obstacle for migrating lattice dislocations. In more recent work by Brons et al. (2013a,b), TB mechanisms, such as detwinning, were observed to coincide with abnormal grain growth observed in nanoindentation of NC Cu thin films at cryogenic temperatures, especially in $\langle 100 \rangle$ textured columnar NC Cu. The evolution of twin density within the various regions of the indented thin film suggests their role in strain accommodation and influence on other nanoscale mechanisms as a function of orientation and resolved stress state.

To further study the role of TBs in face-centered cubic metals, other efforts have utilized molecular dynamics (MD) simulations. Frøseth et al. (2004) used NC Cu and Ni structures to show that the influence of TBs on the deformation mechanisms is not identical for all fcc metals. They explained these differences by analyzing the generalized planar fault (GPF) energy curves for both dislocation nucleation and TB migration. In addition, their simulations show that emitted dislocations prefer to slip on planes of maximum Schmid factor, and TB migration alone is not responsible for strain accommodation in nanotwinned (NT) Cu and Ni. In addition, recent atomistic simulations by Wu et al. (2011) on Cu detail critical dislocation mechanisms as migrating dislocations interact with TBs. Their rationale is based on the fact that TB migration must be accompanied by other strain accommodation mechanisms in NT Cu. Therefore, atomistic calculations were employed to elucidate the critical TB spacing for determining the transition of dominant mechanisms that involve dislocation/TB interactions. Their results showed that a maximum in the strength of NT Cu, as a function of TB spacing, coincides with this mechanism transition. At smaller twin spacings, Li et al. (2010) showed that dislocation nucleation controlled behavior was fundamental to a softening trend in NT metals. Their MD simulations suggest that the strength of NT Cu is dependent on both the average grain size and TB spacing. Furthermore, they conclude that in structures with smaller-spaced TBs, dislocations are nucleated at the TB-GB intersection (due to a local high stress concentration) and glide along the TBs inducing TB migration, as observed earlier by Wang et al. (2007). Therefore, TBs can be preferred nucleation pathways for dislocations and possibly act as sites for further plastic deformation.

These experimental and computational results have led to a significant understanding of the influence of TBs in nano-twinned metals, and have provided insight into the vital structure-property relationships that govern material behavior. However, many conclusions of previous MD simulations about the cooperation/competition of different nanoscale deformation mechanisms, as a result of TBs in the microstructure, have been mostly qualitative. There have been more recent efforts, however, aimed at quantifying plastic deformation within nanocrystalline metals using post-processing methods and atomistic simulations, such as Stukowski et al. (2010) and Vo et al. (2008). In Stukowski et al. (2010), the identification and determination of dislocation and planar fault densities is provided in the context of twinning effects in equiaxed NC Cu and Pd. While in Vo et al. (2008), the strain contribution of dislocations and GB sliding was quan-

tified in equiaxed NC Cu during uniaxial compression as a function of strain rate and grain size. These efforts represent a significant stride in (1) understanding the nanoscale strain accommodation mechanisms in nanocrystalline metals and (2) developing innovative tools for data analysis from atomistic simulations. Inspired by these efforts and the initial findings/questions of [Brons et al. \(2013a\)](#), our objective is to elucidate the influence of grown-in TBs in $\langle 100 \rangle$ textured columnar NC Cu on the active nanoscale deformation mechanisms, especially the interplay of dislocation slip and TB migration, and their role in strain accommodation within the microstructure using large-scale MD simulations and post-processing methods formulated from continuum mechanics theory. To accomplish these objectives, we leverage non-local kinematic metrics and atomistic fields to compute the evolution and interplay of different deformation mechanisms commonly observed within NC fcc metals. In addition, we quantitatively show how TBs and the loading direction influence the role of nanoscale processes in accommodating strain within the microstructure, and provide a new approach for analyzing the deformation fields (especially TB migration volumes) within atomistic simulations.

2. Simulation Methodology

In this study, we utilize three different $\langle 100 \rangle$ textured columnar NC Cu configurations. One structure contains no grown-in TBs (Figure 1(a), 'No Twins'), another contains a single pair of TBs in the middle of each grain (Figure 1(b), 'Single Twins'), and the last one contains multiple TBs per grain (Figure 1(c), 'Twins'). To create the structures, we employed a series of steps. First, three different $\langle 100 \rangle$ cylinders were generated: (1) pure single-crystalline, (2) a single pair of TBs with 2nm spacing, and (3) numerous TBs with 2nm spacing. Each cylinder was approximately 10nm in diameter and 30nm in length. The axial orientation of each cylinder was held fixed in the $\langle 100 \rangle$ direction. Periodic boundary conditions were employed along the axis of the cylinder and an embedded atom method interatomic potential from [Mishin et al. \(2001\)](#) was used. In this work, we studied $\langle 100 \rangle$ columnar NC Cu due to the interesting results of [Brons et al. \(2013a\)](#) and the fact that we wanted to have TBs oriented at an angle to the axis of straining, instead of parallel or perpendicular. Having the TBs oriented at an angle to the loading axis leads to both normal and resolved shear components on the TBs, to aid in their migration (as covered below).

The second step started by creating an amorphous Cu slab with dimensions of approximately 50nm (X) by 50nm (Y) by 30nm (Z). The cylinders generated from step one were then inserted into the amorphous slab at random positions (within the X-Y plane) and with random rotations around the axial direction. The atoms in the amorphous slab that overlapped with atoms in the cylinders were removed. These cylinders with random rotations formed the seeds that induced grain misorientations within the amorphous slab. This step was performed with three separate amorphous slabs, one for each type of cylinder, based on the TB configuration of the cylinder. The random locations and rotations of each cylinder within the amorphous slabs were identical for each of the three structures to generate three similar grain morphologies that differ in their initial TB distribution.

Finally, the structures containing the inserted cylinders (or grain seeds), separated by amorphous regions were allowed to relax and equilibrate to 300K under NPT and three-dimensional periodic boundary conditions. Zero normal pressure was prescribed in each direction, and the thermal equilibration continued for 10 million timesteps, or 10 nanoseconds. After thermal equilibration, as Figure 1 shows, three different columnar NC structures were created containing different initial TB densities in each grain. In each structure there are approximately 6,000,000 atoms and the average grain size is about 14 nm. Note that the crystalline grain seeds have grown until the resulting crystalline grains have impinged on each other and that the initial amorphous regions are no longer present. This approach, unlike most methods using a Voronoi tessellation based procedure, allows for the existence of initial defects within the grains and along the initial TBs, leading to a more realistic starting microstructure. Producing starting microstructures for atomistic simulations that contain non-perfect TBs (i.e., containing defects/steps) has been previously discussed by [Wang et al. \(2013\)](#). Furthermore, as the initial microstructures used in this work contain approximately 14-16 columnar grains (or about 4 grains along each X and Y dimension), it is worthwhile to note the possibility that the mean-field behavior or statistical grain behavior might not be fully captured (as discussed by [Chen and Ngan \(2011\)](#)).

Figure 1: The initial $\langle 100 \rangle$ textured columnar NC Cu configurations containing (a) no TBs ('No Twins'), (b) a single pair of TBs ('Single Twins'), and (c) multiple TBs per grain ('Twins'), where atoms are colored according to their computed CNA value (blue = fcc, aqua = hcp (stacking faults and TBs) and red = other (defects and GBs)).

Each atom in Figure 1 is colored according to the common neighbor analysis (CNA) method (Faken and Jonsson (1994); Tsuzuki et al. (2007)), highlighting the local crystalline structure of each atom based on its local atomic neighborhood. For example, atoms that have 12 nearest neighbors are either face-centered cubic (fcc) or hexagonal close-packed (hcp), while atoms that do not have 12 nearest neighbors are non-12 coordinated atoms. Non-12 coordinated atoms are those atoms located within GBs or other defects. The CNA method offers an advantageous technique to both visualize and distinguish different groups of atoms based on their local crystalline structure. For example, atoms located within stacking faults and TBs are in the hcp group and can be differentiated from other fcc atoms using their computed CNA values. Other useful metrics for determining instantaneous atomic properties, such as potential energy and centrosymmetry, are not used to visualize or group atoms in this study, but are certainly valuable for analyzing atomistic simulations. All simulations performed in this work were carried out using the LAMMPS software package (lammmps.sandia.gov, Plimpton (1995)), while visualizations were created using OVITO (ovito.org, Stukowski (2010)).

After the initial relaxed structures were generated, uniaxial constant strain-rate (10^8 s^{-1}) deformation was performed along the Z-direction (tension and compression), corresponding to the $\langle 100 \rangle$ textured direction of each microstructure. During isothermal straining at 300K, zero normal stress was maintained in transverse directions (X and Y) under NPT. Uniaxial straining continued for 1.5 nanoseconds until 0.15 engineering strain was achieved and significant deformation had occurred within the structures. Snapshots of the atoms during straining were output, containing atomic position, CNA, and the atomic stress tensor. The use of the virial stress tensor and other stress formulations within atomistic simulations has been previously discussed (Zhou (2003); Zimmerman et al. (2004)). Post-processing methods for computing deformation mechanisms, TBs, and twinned volumes used to analyze the simulations, examining the influence of TBs and straining, will be outlined in the following discussion.

2.1. Determining Deformation Mechanisms and Atomic Groups

For analysis, we have both developed and utilized post-processing algorithms to determine the atomic fractions of atoms that have undergone slip due to both partial and perfect dislocations, atoms lying within TBs, and atoms that have been traversed by migrating TBs (i.e., twinned volume). These algorithms rely on computed neighbor lists, atomic CNA values, atomic slip vector, and microrotation from the simulation snapshots. Atomic microrotation, as defined below, computes local non-homogeneous rotation about an atom, given its neighboring atoms and reference/current interatomic vectors. Atomic groups with similar characteristics are identified and tracked during straining within the outlined simulations, such as atoms that have been traversed by a migrating TB or a partial dislocation. Once these groups (or mechanisms) are defined, our analysis shows the resulting evolution of each mechanism during microstructure deformation and the contribution of each mechanism to the overall strain accommodation within the structure. In the

following paragraphs we outline the methodology for computing slip vector, microrotation, and atomic strain from microscale kinematic metrics.

Slip vector (Zimmerman et al. (2001)) for atomistic simulations has commonly been used to estimate the local Burgers vector of each atom, based on its computed neighbor list, due to dislocation slip. The computed atomic slip vector is a function of both the computed neighbor list and the chosen reference configuration to compare with the deformed configuration. In our simulations, we utilize the first nearest neighbor list to compute each atom's slip vector (s^α) and the initial undeformed microstructure at zero strain. To compute s^α , the following equation is used.

$$s^\alpha = -\frac{1}{n_s} \sum_{\beta \neq \alpha}^n (x^{\alpha\beta} - X^{\alpha\beta}) \quad (1)$$

The computed slip vector of atom α (s^α) is a function of the number of first nearest neighbors (n), the number of slipped nearest neighbors (n_s), and each nearest neighbor's (β) distance from atom α in the current ($x^{\alpha\beta}$) and reference ($X^{\alpha\beta}$) configurations. Slip vector estimates the relative slip of each atom's surrounding nearest neighbors with regard to the initial undeformed configuration. For example, atoms located within a stacking fault in fcc Cu, due to slip by a partial dislocation, have a slip vector magnitude approximately equal to the theoretical value for a partial dislocation in Cu (1.48Å), while the computed slip vector for atoms that have been traversed by both a leading and trailing partial dislocation is near the Burgers vector value for a perfect dislocation (2.56Å), assuming an equilibrium lattice parameter value of 3.615Å for Cu.

As stated above, atomic slip vector (s^α) is a useful analysis tool for estimating the relative slip about an atom from atomistic simulations, as compared to an initial reference state or configuration, based on neighbor lists. Additional metrics can also be defined to estimate other kinematic quantities of interest, like atomic microrotation and strain, to probe the history or path of atomic fields due to deformation. Previous studies (Tucker et al. (2011a,b); Zimmerman et al. (2009)) have detailed and shown the utility of various computed microscale kinematic metrics in atomistic simulations. These approaches, similar to computing atomic slip vector, rely on neighbor lists from a defined reference (or initial) atomic configuration. However, the formulation of these microscale kinematic metrics are based on continuum mechanics theory. Here, we briefly outline the procedure used in this manuscript to compute the distribution and resolve the contributions of various nanoscale mechanisms to the overall deformation in textured columnar NC Cu.

Specifically, we review the formulations for the metric tensors of atomic microrotation (ϕ^α) and atomic strain (E^α) for atom α . The reader is encouraged to consult Tucker et al. (2010), Tucker et al. (2011a), and Tucker et al. (2011b) for further details about these atomic metrics. To compute the atomic microrotation tensor (ϕ^α) for atom α , we leverage the computed atomic deformation gradient tensor (F^α) from Zimmerman et al. (2009) as

$$F^\alpha = \left(\sum_{\beta=1}^n (x^{\alpha\beta})(X^{\alpha\beta}) \right) \left(\sum_{\beta=1}^n (X^{\alpha\beta})(X^{\alpha\beta}) \right)^{-1} \quad (2)$$

and the skew-symmetric part of the atomic rotation tensor (R_{skew}^α) from the decomposition of the atomic deformation gradient tensor, as shown in the following equation and outlined in Tucker et al. (2010, 2011b).

$$\phi^\alpha = -\frac{1}{2}\epsilon(R_{skew}^\alpha) \quad (3)$$

where ϵ is the permutation tensor, and the other quantities are identical to those used in Eqn. 1. For these computed atomic metrics, the initial/undeformed atomic configuration was utilized as the reference, along with first nearest neighbor lists. As discussed in Tucker et al. (2010) and Tucker et al. (2011b), additional neighbors can be included in the calculations to alter the non-local consideration of each metric. However, care must be taken in properly weighting the additional neighbor shells.

The atomic microrotation tensor (ϕ^α) is also useful for exploring nanoscale deformation fields and local lattice rotation as outlined by [Tucker et al. \(2011a\)](#) and [Tucker et al. \(2011b\)](#). As the cooperation/competition of different mechanisms in NC metals becomes more important, atomic microrotation can also uncover unique kinematic signatures of the various deformation mechanisms from atomistic simulations, as shown in [Tucker et al. \(2011a\)](#). However, in this work our aim is to elucidate the influence of TBs on the mechanics and deformation of the NC Cu structures. Therefore, as shown in [Tucker et al. \(2011a\)](#), we leverage the formulation of the atomic strain tensor (E^α) to resolve the contribution of each computed mechanism to the overall uniaxial strain of the configuration during the simulations. The atomic strain tensor (E^α) is computed according to the following equation.

$$E^\alpha = \frac{1}{2} [(F^\alpha)^T F^\alpha - I] \quad (4)$$

In this equation, E^α is the atomic strain tensor ([Tucker et al. \(2011a\)](#)), F^α is the atomic deformation gradient tensor, and I is the identity tensor. Throughout the manuscript, we will attempt to differentiate between imposed strain and computed atomic strain by referring to the strain imposed in the simulation cells due to the prescribed constant strain rate conditions applied in these simulations as *imposed strain*, while the strain computed from the atomic strain tensor will be simply referred to as *strain* or *atomic strain*. The same kinematic metrics are used for all structures in this study. In addition, these same metrics can conceivably be used for deformation analysis of other metallic systems and non-crystalline systems as well.

3. Results and Discussion

3.1. Mechanical Behavior and Microstructure Evolution

Uniaxial tensile and compressive straining is imposed at a constant $10^8 s^{-1}$ strain rate on the simulated structures to study the influence of TBs on both the mechanics and deformation of $\langle 100 \rangle$ textured columnar NC Cu. The imposed straining is performed at 300K under NPT along the axial direction (Z) of the columnar NC structures, with zero prescribed normal stress in transverse directions (X and Y). We leverage the computed stress tensor for each atom to generate the stress-strain curves shown in Figure 2. The global stress tensor is computed by summing each atom's stress tensor and normalizing by the total number of atoms. Figure 2(a) shows the uniaxial tension results of the three different columnar microstructures: 'No Twins' (black), 'Single Twins' (blue), and 'Twins' (red), while Figure 2(b) provides the uniaxial compression behavior for the same structures. Each simulation is continued until 15% imposed strain is achieved and significant plastic deformation has occurred within the microstructure, as shown in Figure 3 for the three deformed microstructures at 15% uniaxial tensile strain (a-c) and compressive strain (d-f), where atoms are colored according to the CNA method (fcc = blue, hcp = light blue, and other = red).

The elastic modulus for these structures is obtained from a linear fit of the stress-strain curves in Figure 2 for stresses up to 1.5% imposed strain. The resulting elastic moduli from Figure 2(a) for the three structures under uniaxial tension are 63.1 GPa ('No Twins'), 70.0 GPa ('Single Twins'), and 77.4 GPa ('Twins'). For an ideal $\langle 100 \rangle$ single-crystalline structure (containing no TBs or GBs) for this potential and computed in the same manner, the modulus under tension is 70.4 GPa. Note that there is a modest influence of the TB density on the elastic behavior during tension. However, the initiation of plastic deformation in each structure, based on the peak tensile stress and visual inspection of the structures, shows a clear dependence on the underlying TB density. The large peak in the stress-strain behavior for the 'No Twins' structure indicates that the onset of plastic deformation requires a larger activation stress than the other two structures, and that plastic deformation also occurs at a larger relative strain. The tendency to observe high peak stresses in molecular dynamics simulations (as observed for the 'No Twins' structure) has recently been discussed by [Brandl et al. \(2009\)](#) and is based on the requirement of imposing high strain-rates within atomistic simulations. As a single pair of TBs are introduced (i.e., 'Single Twins' structure), the peak stress is considerably lower and occurs at a lower strain. With numerous TBs introduced into the microstructure (i.e., 'Twins' structure), there is no observable peak in the stress-strain behavior. In fact, the behavior is more representative of polycrystalline samples strained at much lower strain rates than those imposed in these simulations, lacking

an initial large activation stress for inelastic deformation. Following incipient plasticity, the flow stress is similar for each structure. For example, the computed flow stresses between 7% and 15% imposed strain are 1.86 GPa ('No Twins'), 1.84 GPa ('Single Twins'), and 1.94 GPa ('Twins'), indicating negligible influence of the varying microstructures on the flow stress.

Figure 2: The uniaxial (a) tension and (b) compression stress-strain behaviors for the three different microstructures.

Under uniaxial compression (Figure 2(b)), the elastic modulus values are 55.3 GPa ('No Twins'), 61.2 GPa ('Single Twins'), and 70.7 GPa ('Twins') and the corresponding ideal $\langle 100 \rangle$ single-crystalline modulus under compression is 64.7 GPa. There is a similar dependence of the moduli on the microstructure, as observed under uniaxial tension. However, one drastic difference between the stress-strain behaviors under uniaxial tension and compression is the lack of a distinct stress peak during compression straining, coinciding with the onset of plastic deformation, for both the 'No Twins' structure and the 'Single Twins' structure, as seen in Figure 2(a) for tension. For the uniaxial compression results, the initiation of plastic deformation mechanisms occurs without a large activation stress and at lower relative strains than observed during tensile straining. Another difference between the curves is the dependence of the flow stress on the microstructure (or TB density). The computed flow stresses between 7% and 15% strain under compression are slightly lower for each structure than under tension: 1.38 GPa ('No Twins'), 1.45 GPa ('Single Twins'), and 1.59 GPa ('Twins'), indicating that the continual activation of inelastic deformation mechanisms (which will be discussed in detail later) requires lower stresses under compression than tension within the $\langle 100 \rangle$ textured NC Cu microstructures at 300K.

The observed tension-compression asymmetry in the flow stresses of these NC structures is interesting, as compared to previous MD simulations (Lund et al. (2004); Tschopp and McDowell (2007); Tschopp et al. (2008)) of equiaxed NC structures, GBs, and single-crystalline dislocation nucleation, where compression loading generally exhibits greater yield or flow stresses than observed under tension. However, there are important microstructural distinctions that need to be carefully considered and weighed when comparing the findings here with previous studies. Most NC deformation studies investigate equiaxed grains, while the current structure is not equiaxed, it is $\langle 100 \rangle$ columnar (motivated by Brons et al. (2013a)). In nanograined materials, such as NC copper, GBs become increasingly more influential in strain accommodation mechanisms and plasticity, than the grain interiors. However, in the textured columnar structures studied here, we have GBs that are oriented parallel to the loading axis and therefore do not have a significant resolved normal/shear stress state, as true in equiaxed NC structures with GBs oriented at various angles with respect to the loading axis. As such, their role in strain accommodation is limited (discussed and quantified below) and will likely lead to significant differences in the flow-stress tension-compression asymmetry. Also, the texture or grain orientation of the structure plays a crucial role in the deformation response and needs to be considered carefully before direct comparison, as further explained below.

Figure 3: The deformed microstructures at 15% strain for uniaxial tension (a-c) and compression (d-f), for 'No Twins' (left), 'Single Twins' (center), and 'Twins' (right) microstructures - atoms are colored according to their computed CNA values.

Figure 3 shows the resulting deformed microstructures at 15% imposed strain under both uniaxial tension (a-c) and compression (d-f) for each of the microstructures: (left) 'No Twins', (center) 'Single Twins', and (right) 'Twins'. Each atom in Figure 3 is colored according to the CNA method. After straining, significant deformation has occurred in each microstructure and the imposed strain has been accommodated through a variety of mechanisms, including dislocation nucleation/migration, TB nucleation/migration, and GB plasticity. Stacking faults and TBs (light blue atoms) are clearly evident in the deformed structures, as well as partially disordered intercrystalline regions (due to GB deformation). Under uniaxial tension, there is a clear dependence of the initial TB density on the resulting concentration of both stacking faults and TBs. In the 'No Twins' structure (3(a)), the deformed microstructure lacks a significant density of either stacking faults or TBs. The density of both stacking faults and TBs in the deformed structures appears to be a function of the initial TB density under tension, as both the 'Single Twins' and 'Twins' structures contain higher concentrations, as compared to the 'No Twins' structure. Specifically, the TB density in the 'Twins' structure is greater than the other two structures, indicating the stability of initial nanoscale TBs in these columnar NC Cu samples.

Under compression 3(d-f), the resulting defect structures after 15% imposed strain appear to be less influenced by the initial TB density as compared to tension. However, there is still evidence of a slight role of the initial microstructure on the resulting stacking fault and TB densities, as the 'No Twins' structure contains slightly fewer TBs as compared to either the 'Single Twins' or 'Twins' structure. When comparing the resulting deformed microstructures between uniaxial tension and compression, it is also clear that the concentration of both stacking faults and TBs is greater under compression than tension for both the 'No Twins' and 'Single Twins' structures. Once again highlighting the potential role of both the loading direction and initial microstructure on the deformation properties.

One useful technique for estimating the density of different atomic groups, such as atoms located within the fcc lattice, stacking faults, TBs, and other defects (such as GBs) is to compute the evolutionary behavior

of atoms based on the CNA method. This approach has recently been employed in other atomistic simulations [Tucker and Foiles \(2013\)](#); [Tucker et al. \(2011a\)](#). For this study, we focus on three different computed CNA groups: *fcc*, *hcp*, and *other* non-12 coordinated atoms (such as GB atoms). We can sum the total number of atoms in each of these three groups based on each atom's CNA value, normalized by the total number of atoms, and plot as a function of the imposed strain for both tension and compression for each structure. Figure 4 shows the evolutionary behavior of the (a) *fcc*, (b) *hcp*, and (c) *other* atomic groups, where the closed symbols signify uniaxial tension and open symbols are for compression. The black symbols and lines are for the 'No Twins' structure, blue represents the 'Single Twins' structure, and the red symbols and lines for the 'Twins' structure. In each plot, the summed number of atoms in each group, according to the computed CNA value, has been normalized by the total number of atoms in each structure to provide the *Atomic Fraction*.

Figure 4: Evolution of the (a) *fcc*, (b) *hcp*, and (c) *other* (GB/defect) atomic fractions as a function of imposed strain for both uniaxial compression (open) and tension (closed) at room temperature.

As Figure 4(a) shows, the fraction of *fcc* atoms continues to decrease with deformation and imposed straining for each structure under both tension and compression. However, as qualitatively noted earlier, there appears to be a slightly larger influence of the initial microstructure (i.e., TB density) under tension than under compression. This trend is shown in the evolution of the *fcc* atomic group in Figure 4(a). There is less variance in the resulting atomic fraction of *fcc* atoms for each structure under compression (open symbols) than under tension (closed symbols). In addition, notice that there is less difference between tension and compression behavior for the 'Twins' structure, indicating once again the influential role of the initial TB density on the resulting microstructural evolution during imposed straining.

The evolution of the *hcp* atomic group is shown in Figure 4(b), and note that this atomic group contains atoms located within both stacking faults and TBs. There is a continuous increase in the *hcp* atomic fraction with deformation, as both stacking faults and TBs are produced during straining. The production of stacking faults in these NC simulations is linked to the nucleation of partial dislocations within fcc metals. As observed for the *fcc* atomic group, there is less variance in the resulting *hcp* atomic fraction under compression, than under tension, indicating both the role of the initial microstructure and the applied loading conditions. In addition, for both the 'No Twins' and 'Single Twins' structures, there is a greater fraction of *hcp* atoms under compression than tension, supporting earlier observations of the deformed microstructures. Furthermore, the difference in the atomic fraction of the *hcp* atomic group for the 'Twins' structure under compression and tension is negligible, similar to the *fcc* atomic group shown in 4(a).

This behavior does not continue for the *other* atomic group in the 'Twins' structure, as displayed in Figure 4(c). Once again, this atomic group contains mostly GB atoms, but also includes atoms within other defects structures, like dislocation cores and TB steps. For both tension and compression, the atomic fraction of *other* atoms continues to increase with imposed strain for the 'No Twins' and 'Single Twins' structures; however, there is a difference in the 'Twins' structure. Under tension, the atomic fraction of the *other* atomic group continues to rise with imposed strain, while there is a decrease at higher imposed compressive strain. However, the total atomic fraction difference between tension and compression for the *other* atomic group is

only about 1%, while differences in the *fcc* and *hcp* atomic groups for the 'Twins' structure are approximately 2% and 1%, respectively. These comparisons show that detailed differences between microstructural features, and possibly the active deformation mechanisms, are difficult to ascertain using commonly used metrics. Furthermore, this technique also points to the need for more tailored methods for differentiating between strain accommodation mechanisms during plastic deformation in NC systems. In particular, we will focus in this study on differentiating between dislocation slip and TB nucleation/migration, highlighting how the *hcp* atomic group can be subdivided into both stacking faults and TBs.

3.2. Distribution of Deformation Mechanisms

During deformation, our atomistic simulations show a variety of deformation mechanisms, such as dislocation slip, TB nucleation/migration, and GB plasticity. In this section, we will focus on differentiating between dislocation and TB mechanisms during deformation and discuss a newly developed technique for computing the atomic fraction of atoms participating in partial dislocation slip, perfect dislocation slip, TB nucleation, and TB migration. These distinctions will highlight the role of both the initial TB density and the loading conditions on the evolution of plastic deformation mechanisms within these NC columnar Cu samples.

Figure 5: Images showing vertical slices through the columnar microstructures that highlight the influence of TBs on the dislocation activity during tension (top, a & b) and compression (bottom, c & d) for the 'No Twins' (left, a & c) and 'Twins' (right, b & d) microstructures. Perfect dislocation glide planes are highlighted red, GB/defect atoms are gold, *hcp* atoms are light blue, and remaining *fcc* atoms are blue.

Dislocation nucleation and migration are observed during both uniaxial tension and compression in all three structures. However, the slip behavior of dislocations being emitted from GBs are significantly influenced by the initial TB density. Partial dislocations are emitted from GB regions during deformation and travel across the grain until they interact with other defect structures, such as GBs, stacking faults, and

TBs. Trailing each partial dislocation is a stacking fault that is terminated at either its originating GB or a trailing partial dislocation. Recent publications have discussed the effect of the generalized stacking fault energy curve and twinning barrier for various fcc metals to nucleate leading/trailing partial dislocations and twins (Van Swygenhoven et al. (2004); Weinberger and Cai (2012)); therefore, we will not discuss these aspects in this work. Here, we refer to atoms lying within stacking faults as 'partially slipped', meaning that their computed slip vector should approximate that of the Burgers vector for partial slip, as discussed earlier, while atoms that have been traversed by both a leading and trailing partial dislocation have been 'perfectly slipped'.

Figure 5 shows how both the initial TB density and loading direction influence the resulting dislocation activity at 10% imposed strain under tension (a-b) and compression (c-d). To identify 'perfectly slipped' atoms, we use the atomic slip vector (s^α) and CNA value for each atom. A perfectly slipped atom will be in the *fcc* atomic group and have a computed slip vector magnitude near the theoretical Burgers vector value for perfect dislocation slip, as discussed earlier. The computed slip vector is what allows us to differentiate perfectly slipped atoms from other non-slipped *fcc* atoms. In Figure 5, perfectly slipped atoms are colored red, all other *fcc* atoms are blue, *hcp* atoms are light blue, and GB/defect atoms are gold. In the 'No Twins' structure (5(a) and (c)), multiple perfect dislocation glide planes are observed at different orientations with respect to the loading axis. Perfect dislocations are seen to interact and cross numerous stacking faults and TBs. However, in the 'Twins' structure (5(b) and (d)), the higher density of TBs seems to limit the activation of many dislocation glide planes not parallel to the TBs. Perfect dislocations tend to glide parallel to the TBs in each grain and few high-angle dislocation/TB interactions are observed, as was seen in the 'No Twins' structure. This behavior is consistent throughout the microstructure, and it appears from Figure 5 that *dislocation activity in the 'Twins' structure is suppressed, as compared to the 'No Twins' structure*. The simulation results shown in Figure 5 suggest that TBs could act as obstacles to dislocation motion, TBs could be favored pathways for dislocations nucleated from TB-GB intersections (in agreement with Wang et al. (2007)), and the activation of alternative deformation mechanisms could be linked to a higher TB density. However, a more detailed comparison is needed to uncover the exact role of TBs on the activation of different mechanisms.

Figure 6: The evolution of both (a) partial and (b) perfect dislocations during straining for uniaxial tension (closed) and compression (open).

First, we compute and compare the evolution of both partial and perfect slip during deformation. The computed atomic slip vector and each atom's CNA value will be leveraged. Partially slipped atoms will have a slip vector magnitude approximately equal to the theoretical Burgers vector magnitude for partial slip and be in the *hcp* atomic group. Once we compute both partially and perfectly slipped atoms we can plot the *Atomic Fraction* of each group as a function of imposed strain, as shown in Figure 6. Figure 6(a)

shows the evolution of partially slipped atoms due to partial lattice dislocation migration and Figure 6(b) shows the evolution of perfectly slipped atoms due to the migration of perfect lattice dislocations for both compression (open symbols) and tension (closed symbols) for each of the three NC structures.

Under both tension and compression, the fraction of partially slipped atoms tends to level off during further imposed straining, while the fraction of perfectly slipped atoms continues to increase. However, the fraction of perfectly slipped atoms is greater in tension than in compression, and partial dislocations tend to be more prevalent in compression than in tension. The data also show that initial TB density is influential on the resulting dislocation activity, as the perfect dislocation activity is greater in the 'No Twins' structure than the other two structures. However, in general, lattice dislocation activity is less significant under compression than in tension, and less significant for the 'Twins' structure than the other structures, indicating that other deformation mechanisms must be more active under compression and in the more highly twinned (i.e., 'Twins') structure.

Figure 7: The evolution of (a) TBs and (b) twinned volume for uniaxial compression (open) and tension (closed) simulations.

To further explore this issue, we leverage a new approach, based on local atomic neighborhoods, to calculate the evolution of TBs during deformation and possibly capture any tendency to nucleate new TBs from the untwinned ccc lattice. To compute the atomic fraction of atoms lying within TBs, our algorithm computes the nearest neighbor list for each atom and iteratively searches for neighbors within the *hcp* atomic group. Therefore, an atom lying within a TB must be in the *hcp* atomic group and have exactly 6 nearest neighbors also within the *hcp* group. One caveat, however, is that the edges of stacking faults can be included in this calculation, and the edges of TBs will not be included, but we have performed extensive calculations to verify the accuracy of this approach, and we found an error of less than 1% in these simulations.

As a check to this approach, we also employed a more computational intensive calculation that verifies that the nearest *hcp* neighbors were planar, as in a TB. This approach led to almost identical atomic fractions for TB atoms, and was much more computationally expensive. The resulting evolution of the fraction of atoms within TBs is shown in Figure 7(a). These results agree with the observations of the deformed microstructures shown in Figure 3. During compression, the fraction of atoms lying within TBs is much greater than under tension at higher imposed strains for the 'No Twins' and 'Single Twins' structures. Additionally, the nucleation of new TBs does not occur until about 4% imposed strain in both the 'No Twins' and 'Single Twins' structures. However, the atomic fraction of TBs within the microstructures under both uniaxial tension and compression were almost identical for the 'Twins' structure, indicating that the creation of new TBs (i.e., TB nucleation) alone does not make up for the lack of dislocation activity seen in the 'Twins' structure, as discussed above.

During straining, we noted significant differences in the resulting TB distributions within the 'Twins'

structure, when comparing tension and compression results. It appeared as though the propensity for TB migration differed for each structure and loading direction. Therefore, we implemented another post-processing technique for computing the volumes within the microstructure that have been traversed by a migrating TB, we call these volumes 'twinned volumes'. This method relies on the computed neighbor lists, each atom's CNA value, and atomic microrotation as outlined in Section 2. The microrotation metric is a dual vector representation of the skew-symmetric part of the atomic rotation tensor. In other words, it captures the non-uniform rotational deformation field around an atom as compared to the initial (undeformed) configuration and atomic neighborhood. So, as a TB migrates, due to the propagation of twinning partial dislocations along its plane, atoms that have been twinned are *fcc* and have a microrotation magnitude greater than a specified lower limit of 0.1 using our formulation. In our simulations, we used pristine twinned nanowires to validate this microrotation lower limit to capture twinned volume atoms. Figure 7(b) shows the resulting evolution of twinned volume atomic fraction for each structure under both uniaxial tension and compression.

Figure 7(b) shows that TB migration is more prevalent in the 'Twins' structure than the other structures, and greater under compression than tension in all three microstructures. Clearly, both the initial density of TBs and the loading direction influence the propensity for TB nucleation and migration, as captured in Figure 7. These results also indicate that TB migration is a considerable strain accommodation mechanism in $\langle 100 \rangle$ NC columnar Cu microstructures with a high density of TBs and nanograins, and specifically under compressive loading conditions. It is likely that both twinning (through TB nucleation) and detwinning (through TB migration and the annihilation of existing TBs) occur simultaneously in some of these structures during loading. The migration of a TB plane, as observed in our simulations, occurs via the motion of a single partial dislocation adjacent to an existing TB plane (as termed a twinning partial dislocation), while the motion of a single partial dislocation within the lattice creates a stacking fault identified as two adjacent *hcp* atomic planes. TB nucleation, or the creation of new TB planes (identified as a single *hcp* atomic plane, on the other hand, occurs when two partial dislocations slip on neighboring glide planes. Each of these mechanisms are observed in our simulations, and our metrics capture twinning partial dislocation migration within the partial dislocation group.

Since the presence of TB migration increases with a high density of TBs, our results indicate that the TBs tend to act as preferred nucleation pathways for dislocations emitted from the GBs at the intersection of the TB and GB. The activation of TB migration (through the migration of twinning partials along the TB plane) seems to compensate for the lack of both lattice dislocation activity and TB nucleation in the 'Twins' microstructure, and under compression in general. The disparity between partial/perfect dislocations and TB plasticity during uniaxial straining can possibly be explained by the non-glide (i.e., normal) resolved stresses on the slip/twin planes, as explained by Lund et al. (2004) for small-grained NC Ni. However, a more likely physical explanation for this mechanistic disparity can be made in regard to the Schmid factors of the partial dislocations, as studied by Jennings et al. (2013).

The work of Jennings et al. (2013) suggests that according to the Schmid factor analysis of partial dislocations, there should indeed be a tension-compression asymmetry in the tendency for partial dislocation nucleation under $\langle 100 \rangle$ loading, where partial dislocation nucleation is key for both lattice dislocation slip (leading and trailing partial dislocations) and TB migration (twinning partial dislocation along TB plane). They found that there is a direction dependence in the calculated Schmid factors for leading and trailing partial dislocations that explains the tendency to nucleate either partial dislocations or perfect dislocations. As it relates to the simulations of this work, we observe that under uniaxial tension, perfect dislocation activity is preferred, while under compression, partial dislocation and twinning activity is preferred. Therefore, as highlighted by Jennings et al. (2013), the Schmid factor for the leading partial dislocation under $\langle 100 \rangle$ tensile loading is lower than the Schmid factor for the trailing partial dislocation, indicating that the nucleation of the trailing partial dislocation should occur immediately following the leading partial dislocation due to a lower energetic barrier for the trailing partial. The partial dislocation ordering is reversed under $\langle 100 \rangle$ compressive loading, where the energetic barrier for the trailing partial is greater (lower Schmid factor) than that for the leading partial, leading to a higher fraction of partial dislocations and twinning activity. The Schmid factor description agrees well with and seems to explain the deformation mechanistic tension-compression asymmetry we observe in regard to dislocation and TB plasticity. The simulation re-

sults provided clearly suggest that TB migration can be a significant mechanism in twinned NC fcc metals (especially under compression loading), but its role in macroscopic strain accommodation is still unclear.

Figure 8: Images showing slices through the microstructures that highlight the resulting atomic volumes (red) that have been traversed by a migrating TB (i.e., twinned) during uniaxial compression for the (a) 'No Twins' and (b) 'Twins' microstructures. The arrows are pointing to the twinned regions. Other atoms are colored according to their calculated deformation mechanism group: partial slip (light blue), perfect slip (green), TBs (yellow), and all other atoms are dark blue.

For clarity, Figure 8 shows the calculated twinned volumes (red) for both the (a) 'No Twins' and (b) 'Twins' microstructures at 10% compressive strain. Arrows are used to highlight the twinned volumes. Atoms in Figure 8 are colored according to their computed deformation mechanism group as follows: partially slipped atoms are light blue, perfectly slipped atoms are green (compare to red atoms in Figure 5(c-d)), TB atoms are yellow, and all other atoms are dark blue. The resulting distinctions shown in this figure display the utility of differentiating different deformation mechanisms within atomistic simulations using non-local atomic field calculations. Additional calculations can now be performed on each deformation mechanism group based on this differentiation. As we discuss next, we leverage this method to resolve the strain contribution of each mechanism to the overall strain accommodation within each microstructure during imposed straining.

3.3. Resolving the Strain Contribution

Now that we have differentiated the deformation mechanisms using the aforementioned method, we can study the contribution of each mechanism to the overall strain accommodation within each microstructure as a function of TB density and loading direction. Figure 9 displays the computed *Fraction of Strain* for each of the atomic groups (i.e., mechanisms) as a function of imposed strain. The *Fraction of Strain* is calculated by summing the computed atomic strain in the loading axis direction (E_{33}) for all atoms in each mechanism group, and then normalizing by the total uniaxial atomic strain for all atoms within the structure. The four groups, or mechanisms, that are included in this study are (1) lattice strain or the *FCC* group, (2) the *Dislocation* group that includes both partially and perfectly slipped atoms, (3) the *TB* group including atoms in both TBs and twinned volumes, and (4) the *GB* group that includes all non-12 coordinated atoms.

In Figure 9(a), the strain contributions within the 'No Twins' structure are shown, where open symbols are for compression and closed symbols for tension. Initially, lattice strain or the *FCC* group, is dominant in accommodating strain, but with further imposed straining, this elastic strain contribution continues to decrease, especially under compression as other mechanisms become more significant. At larger imposed strains, the role of dislocation slip and TBs begins to takeover, as shown in Figure 9(a). Dislocation strain remains higher under tension than in compression throughout the simulations, but strain due to TB migration continues to increase under compression. The strain contribution from dislocation activity is greater than TB activity, and points to the potential role of TBs in altering the activated deformation mechanisms. Furthermore, this figure also shows that the role of GBs in accommodating strain remains low throughout imposed straining. In this work, we do not distinguish between the various mechanisms that would be included in the calculated GB strain, such as GB sliding, atomic shuffling, free volume migration, etc.

Figure 9: The fraction of total computed strain (E_{33}) along the uniaxial direction for the (a) 'No Twins' and (b) 'Twins' structures under compression (open) and tension (closed) showing the evolutionary behavior of *fcc* (black), *dislocation* (blue), *TB* (red), and *GB* (green) strain. Atoms within the *fcc* group do not include atoms in one of the other mechanism groups (e.g., *dislocation* or *TB*).

The distribution of strain within the microstructure is significantly different when a higher density of initial TBs are present, highlighting the potential influence of TBs on the activated mechanisms within NC structures. Figure 9(b) shows the strain behaviors for the different mechanisms in the 'Twins' structure. Similar to the 'No Twins' structure, lattice strain dominates at lower imposed strains, but gives way to other mechanisms at higher strains. In this microstructure, TB migration plays a significant role (especially at higher strains) under both tension and compression, but is certainly more prominent under compression. Strain due to dislocation slip remains low, similar to GB mechanisms. It is worthwhile to note the decreased role of GBs under compression, as compared to tension. This behavior is likely a function of the increased role of the TBs as preferred dislocation pathways under compression. However, unlike the behavior observed in the 'No Twins' structure, strain accommodation due to TB processes is more significant than for dislocation processes. This trend is true for both tension and compression. In addition, strain due to dislocation slip is greater in tension, and strain due to TB nucleation/migration is greater in compression for both structures (i.e., independent of twin density). This indicates that despite the influential role of the microstructure, the loading direction (or resolved stress state) is critically important as well in determining the strain accommodating deformation mechanisms.

Finally, Figure 10 shows images of the microstructure (as shown in Figure 5(c-d) and Figure 8), where atoms are colored according to their computed atomic strain, using first nearest neighbors and the initial undeformed configuration as reference. Included in Figure 10 is a color legend indicating that red atoms correspond to compressive strain, blue atoms are tensile strain, and green atoms are zero strain. Figure 10(a) shows the 'No Twins' structure and (b) shows the 'Twins' structure at 10% compressive strain. In each image there are obvious compressive strain regions highlighted red associated with both twinned volumes and dislocation glide planes. However, dislocation glide planes appear to accommodate the majority of compressive strain in the 'No Twins' structure (in agreement with Figure 9(a)), while twinned volumes in the 'Twins' structure tend to accommodate the majority of compressive strain in the 'Twins' structure. In addition, this figure also shows the comparative strain accommodation due to different deformation mechanisms within the same structure. For example, in both structures, the average magnitude of atomic strain associated with TB migration (or within twinned volumes) is greater than that for both perfect and partial dislocation slip. Furthermore, these images also show the limited strain accommodation within both GB regions and the lattice, also agreeing with the behaviors shown in Figure 9. The calculation of atomic strain, as compared to the reference undeformed configuration, suggest which atomic groups

Figure 10: Images showing slices through the microstructures of the (a) 'No Twins' and (b) 'Twins' structures under compression at approximately 10% imposed strain. Atoms are colored according to their computed atomic strain, as outlined in section 2, where green is no strain, red is compressive, and blue is tensile. These images correspond to the same images shown in Figure 5(c-d) and Figure 8 for comparison.

or mechanisms are more influential in accommodating the imposed uniaxial strain. As the mechanisms of dislocation slip, TB nucleation, and TB migration collectively cooperate to accommodate the imposed strain in these simulations, the calculation of atomic strain for various deformed microstructures provides a valuable perspective and insight into nanoscale strain accommodation and dominant plastic deformation modes in NC metals.

4. Summary

This work has studied the influence of initial nanoscale TBs on the deformation of $\langle 100 \rangle$ columnar NC Cu structures under uniaxial loading at 300K with molecular dynamics simulations. Three different microstructures were created and compared based on their initial TB configurations. In addition, we employed non-local kinematic metrics, formulated for atomistic systems, and neighbor lists to compute microstructure evolution based on different deformation mechanisms as a function of both the initial structure and loading direction. We also developed a new method for identifying regions that have been traversed by a migrating TB and differentiated this mechanism from other mechanisms commonly observed in NC systems, such as dislocation nucleation/migration, TB nucleation, and GB plasticity. By using the defined microscale atomic metrics, we were able to compute the distribution and evolution of different deformation mechanisms that were activated during plastic deformation, and resolve their contribution to the overall strain accommodation within each NC structure. The key findings are summarized below.

In $\langle 100 \rangle$ textured columnar NC Cu, the flow strength is slightly greater under tension than compression, and the presence of initial TBs has a negligible influence on both the modulus and flow strength in these nanograined systems. However, drastic differences emerge when studying the processes fundamental to microstructural evolution. For example, we highlight in this study the differences in the atomic fraction of atoms participating in either partial or perfect dislocation slip during imposed straining. Our results showed that perfect dislocation slip tends to be more prominent than partial slip, and dislocation activity as a whole is less significant under compression than tension. In addition, dislocations were observed to glide parallel to the TB planes in the heavily twinned structure (i.e., 'Twins'), often resulting in TB migration from twinning partials. The 'Twins' structure was also shown to exhibit much lower dislocation activity than the other two structures, so we utilized new methods for estimating TB nucleation and migration.

The simulations showed that TB activity (i.e., nucleation and migration) also displayed significant differences under uniaxial tension and compression. In both the 'No Twins' and 'Single Twins' structures, the TB density increased during compressive straining due to the nucleation of new TBs, but not during tension. However, no differences in the overall fraction of atoms within TBs was observed in the 'Twins' structure during either tension or compression. Therefore, we studied TB migration as a fundamental deformation mechanism using atomistic estimations of microrotation. These results showed that we can identify

regions that have been traversed by a migrating TB using the microrotation metric. In doing so, we were able to quantitatively show that TB migration is indeed a mechanism that is significant in heavily twinned microstructures, and possibly explains the lack of new TBs and dislocation activity. In each structure, TB migration was more significant under compression than tension highlighting the importance of the resolved stress states within the microstructure.

Finally, we used our analysis of the distribution of deformation mechanisms, and computed the fraction of total strain associated with each mechanism using the atomic strain metric. The results show that in the untwinned structure (i.e., 'No Twins'), dislocation slip is the primary strain accommodation mechanism at higher strains, and is greater under tension than compression. While in the twinned structure (i.e., 'Twins'), strain accommodation due to TB migration is the greatest strain accommodation mechanism. These results quantitatively show that TBs do indeed influence the deformation of $\langle 100 \rangle$ columnar NC Cu by altering the activated mechanisms during microstructural evolution, and we can resolve the distribution, evolution, and strain contribution of these different mechanisms by employing kinematic metrics for atomistic analysis.

Future considerations or extensions of the current work can be undertaken to investigate the role of temperature, grain size, GB character/distribution, texture, and TB spacing on the microstructural effects studied here. As previously noted, earlier studies have shown an inherent link between TB spacing and macroscopic material response in twinned fcc metals. However, the analysis methods outlined in this study could greatly contribute to these earlier results by providing numerical evidence for the distribution of the underlying strain accommodation mechanisms. Furthermore, additional straining conditions, such as shear, nano-indentation, and cyclic deformation would further highlight the fundamental role of TBs in the plasticity of NC materials, and potentially aid in the development of materials with tailored mechanical properties.

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