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Correlating deformation mechanisms with X-ray diffraction phenomena in nanocrystalline metals using atomistic simulations

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ABSTRACT

Virtual X-ray diffraction (XRD) and atomistic simulations are used to probe the relationship between XRD phenomena and the strain accommodation methods associated with deformation. Twinned and untwinned nanocrystalline (NC) samples loaded in tension reveal distinct XRD responses. In initially untwinned samples, peak splitting occurs precisely as dislocation mediated deformation mechanisms initiate at approximately 2.9% strain. However, initially pre-twinned samples reveal less dislocation mediated deformation and no observable XRD peak splitting. XRD responses from control sets of ideal defect structures representing bulk and unloaded NC samples are analyzed. This study shows that the peak splitting during deformation of the initially untwinned NC sample can be traced to both the high density of planar defects and the complex internal strain state present under external load.

1. Introduction

Interest in nanocrystalline (NC) materials across the scientific and engineering community has been largely driven by the opportunity for improving mechanical properties, such as increased yield strength, hardness and Young's modulus, as compared to larger-grained polycrystalline materials [1,2]. Fundamentally, the higher density of grain boundaries (GBs) in NC materials is the driving factor for these remarkable improvements. From a mechanistic point of view, GBs can act as significant barriers for dislocation motion, thus limiting the effect of dislocation-mediated strain accommodation in favor of GB-mediated behavior as the average grain size is reduced [3,4]. While the high GB content can improve mechanical properties, it can also have negative effects such as embrittlement and loss of ductility [2,1,5]. Another critical behavior observed in NC materials is property degradation from microstructural instability [6–9]. The high interface area to volume ratio of nanoscale grains and higher energy due to GBs promotes rapid grain growth, diminishing the advantageous properties associated with increased GB density. To counter this process, researchers have focused on stabilizing the microstructural grain morphology through the addition of coherent interfaces (i.e., twin boundaries) to the microstructure [10,2,11,1,12].

In polycrystalline metals, twin boundaries have been observed to act as a barrier for dislocation motion and hinder GB migration [13–15]. Research efforts in the area of GB engineering (GBE) have proposed that the fundamental influence of twins on both dislocations and GB motion can be leveraged to reduce the effects of embrittlement and Ostwald ripening observed in NC metals [16–19]. Since twinning and defect (e.g., dislocation, GB) evolution have been strongly linked to the properties of NC metals, it is critical that researchers have non-destructive means to characterize the internal structure of a sample. To this effect, X-ray diffraction (XRD) techniques are often used as an efficient, non-destructive means of characterizing the microstructure of crystalline samples.

High defect concentration can change a material's internal strain fields resulting in altered XRD profiles as compared to relatively defect-free materials [20–23]. Specifically, defect content can cause both full width at half max (FWHM) broadening [22,20] and peak shifting [22] as measured by Gaussian analysis of isolated peaks and line profile analysis. Ungar et al. [24,25] summarized these differences in 2001, stating that defects such as stacking faults can alter isolated peaks through FWHM peak broadening, asymmetry, and shifting. Based on these early suggestions, Ungar and coworkers [25,24] further stated that XRD can be used to determine several important factors associated with crystalline structure and material properties: the dislocation density, GB volume fraction, the population of Burgers vectors in different slip systems, dislocation character, stacking fault density, and the grain size distribution.

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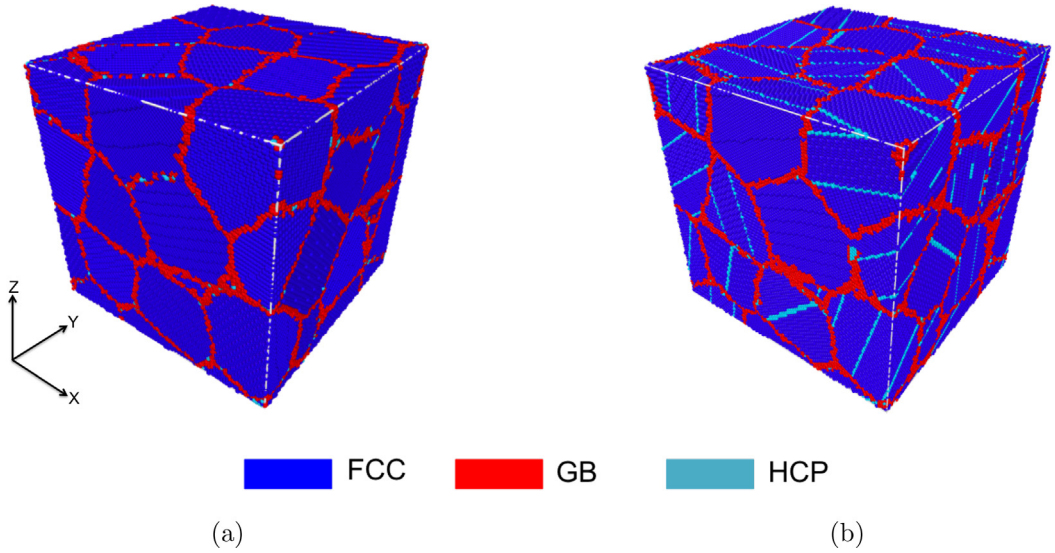

Fig. 1. Three-dimensional microstructures of NC Cu with an average grain size of 10 nm and 29 equiaxed grains, comparing (a) untwinned and (b) twinned grains. Both the (a) untwinned and (b) twinned microstructures have the same grain morphologies and initial grain orientations, but the twinned microstructure contains grains that have been populated with nanoscale twins at 6 nm spacing. Atoms are colored according to their crystalline structure through the common neighbor analysis method (CNA) [46]: blue are FCC atoms, aqua are hexagonal close-packed (HCP) atoms, and red are defect atoms (e.g., GBs). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

The capability of XRD to identify and quantify a multitude of crystalline defects, which are related to the deformation processes of NC systems, make it an intriguing technique for use in the analysis of NC materials [26–28,23,29–31]. Virtual XRD from atomistic models have been used to understand microstrain in NC samples due to lattice dislocations (dislocations not associated with a GB) and other defect content using Williamson–Hall analysis [29–31]. Brandstetter et al. [29] showed that simulated XRD spectra with introduced dislocations and twin content in unloaded NC samples produce similar integral width anisotropy as a function of scattering angle as found in NC experiments. This anisotropy was observed in NC models containing lattice dislocations and was also observed when these lattice dislocations were absorbed into the GBs, due to their associated long-range strain field. Similarly, Markmann et al. [30] showed that microstrain calculated from XRD increases significantly during the deformation of NC samples not as a result of lattice dislocations, but as a result of the long-range strain fields from the heterogenous elastic response and stress concentration at GBs and triple junctions. Using measures of local atomic strain, Stukowski et al. [31] further showed that Williamson–Hall computed microstrain stems from long-range strain fields created by GB dislocations and NC packing constraints rather than atomic relaxations near the GB. More recently, line profile analysis techniques have been shown to extract additional insights. Leonardi and Bish [23] showed that line profile analysis techniques accurately determined defect content and they used simulated XRD to compute the Debye–Waller coefficient for the NC sample at temperature.

Atomistic simulations are a powerful set of tools for systematically exploring how particular defects and internal strain fields alter XRD peak profiles in a manner intractable by experiment. The current work, expands upon the simulation studies by applying virtual XRD techniques developed by Coleman et al. [32] on molecular dynamics simulations of NC Cu deformation. As XRD profiles are computed, strain accommodation mechanisms are identified by microscale kinematic metrics developed by Zimmerman et al. [33] and Tucker et al. [34,17]. This study focuses on understanding the effects that initial twin content has on the underlying atomic deformation mechanisms and shows how these influence the resulting diffraction profiles. Samples that contained twins before deformation showed improved mechanical properties due to the twins acting as additional barriers for defect formation as compared to the untwinned samples. The high density of closely-spaced stacking faults that formed within the untwinned samples combined with the complex strain state during deformation caused a splitting of the simulated select XRD peaks, which is confirmed by examining simulations of different cases of ideal defects.

2. Methodology

2.1. Atomistic simulations

To probe the in-situ characterization capabilities of XRD, molecular dynamics and virtual XRD were used to quantify the influence of the initial twin content on the mechanical behavior, nanoscale deformation mechanisms, and calculated XRD profiles in NC Cu. Two NC samples (i.e., untwinned and twinned) were constructed containing 29 grains with an average grain diameter of 10 nm. The simulation cells were approximately 25 nm × 25 nm × 25 nm and contain approximately 1.5 million atoms. The algorithm used to construct the nanocrystalline atomistic structures avoided unrealistic, needle-like grains by incorporating spherical shape constraints to the Voronoi tessellations as described in Foley et al. [35]. Once the grain centers and average grain size were chosen, atom positions within in each grain were determined by randomly orienting an atomic reference structure. Defect-free grains were thus created by populating the grain volume with a face-centered cubic (FCC) Cu reference, whereas twinned grains were seeded with Cu reference structures containing repeating Cu $\Sigma3\{1\,1\,1\}$ twin boundaries spaced approximately 6 nm apart.

The aforementioned NC generation approach preserved grain morphology for different starting microstructures (e.g., single-crystalline vs. twinned) throughout the study by building each nanocrystalline structure from identical grain centers and grain orientations. This is demonstrated in Fig. 1, showing a single-crystalline (i.e., untwinned) microstructure next to a microstructure containing 100% initially twinned grains.

All atomistic simulations were performed using the LAMMPS [36] simulation package employing an embedded atom method (EAM) [37,38] interatomic potential for Cu developed by Mishin et al. [39]. Prior to deformation, each NC structure was relaxed using a nonlinear conjugate gradient energy minimization followed by a 10 ns thermal equilibration at 100 K under the isothermal-isobaric (i.e., NPT) ensemble with zero normal external stress in all three dimensions. This