PLASTICITY EVOLUTION IN NANOSCALE Cu/Nb SINGLE-CRYSTAL MULTILAYERS AS REVEALED BY SYNCHROTRON X-RAY MICRODIFFRACTION

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Abstract

In this study, the evolution of dislocation densities during compressive deformation of nanoscale Cu/Nb single crystal multilayers with individual layer thickness of 20 nm is investigated using Synchrotron X-ray micro-diffraction. The samples were subjected to successive compression straining up to a final cumulative strain of 35%. The nanolayer composite exhibited a maximum flow strength of ~ 1.6 GPa at approximately 24% compressive strain. Synchrotron X-ray micro-diffraction experiments, using a monochromatic beam of 10 keV energy were performed after each compression strain increment. We observed a significant increase in X-ray ring width peak broadening in both Cu and Nb layers up to strains of $\sim 3.5\%$ followed by saturation broadening at higher strains. This observation indicates that the interfaces of the Cu/Nb nanolayers are very effective in trapping and annihilating dislocation content during mechanical deformation.

Keywords: *Single crystal, Nanolayers, Incoherent interface, Pillar compression, Peak broadening, Dislocation saturation.*

1 Introduction

The next generation of energy systems have attracted serious research attention towards design of multi-functional materials, which can be subjected to extreme environments [1–3]. Among these, nanoscale multilayer composites of Cu/Nb are being keenly explored as model systems to understand the mechanisms that enable ultrahigh strength (~ 2.5 GPa) without compromising its deformability and plastic flow stability ($\sim 30\%$) [4]. This idea of using

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nanolayers to limit the operation of Frank-Read sources within a single layer to build a strong solid was proposed by Koehler [5]. The ability to reach near theoretical strength and large plastic deformation has made nanocomposite materials a serious contender for applications involving high radiation damage, temperature and mechanical loading. The strength of these nanoscale materials are strongly derived from the interface structure compared to its bulk counterpart [6–9]. Hirth [10] also reported the role of interfaces and its effects on mechanical properties of metals. The underlying strengthening mechanisms at this incoherent (*large lattice parameter mismatch*) interface of the face centered cubic (*FCC*) Cu, and body centered cubic (*BCC*) Nb structure, has been investigated using experimental techniques (*ex/in-situ*) [11–13, 4, 14] and simulations (*continuum/atomistic*) [15–26]. Due to the recent report on synthesis of epitaxial Cu/Nb multilayer films where individual layers are single crystalline [27], Synchrotron XRD techniques may be used to probe the evolution of plasticity.

The large strain deformation behavior of the rolled nanoscale Cu/Nb multilayer (each layer of 75 nm thickness) foils was reported by Misra et al. [28]. The transmission electron microscopy (*TEM*) observations of the samples revealed insignificant dislocation cell structure formation and rotations of the interface normal as compared to the meso-scale (each layer of 4 μm thickness) multilayers [6]. They concluded that the plastic flow stability of nanoscale multilayers was a consequence of confined layer slip of single dislocation loops with symmetric slip activity on different systems [29].

To elucidate quantitatively the interfacial strength of the Cu/Nb nanolayers, ex-situ pillar compression and in-situ indentation tests were performed

on these samples by Li et al. [4]. They reported the shear strength of interface to be in the range of 0.3-0.55 GPa. In-situ nano-indentation of these samples were also performed by Li et al. [14] to observe the confined layer slip phenomenon. Their conclusions on these Cu/Nb nanolayers agree well with the insights of atomistic simulations which shows that, due to incoherent interface being weak in shear, they act as potent dislocation traps, via core spreading along interfaces [15, 30, 31]. In our earlier work [13], we probed the overall Laue diffraction peak broadening which is a measure of plasticity of these nanoscale Cu/Nb multilayers. This study reported an increase of statistically stored dislocations in Cu nanolayer, while Nb showed negligible increase at a plastic strain of 14%. A possible crystal recovery of the Nb nanolayer was predicted for this zero net plastic deformation. Based on this study, which showed possible crystal defect annihilations, we further investigate the Cu/Nb single crystal nanolayers to study the plasticity evolution, which is the subject of this paper. The samples used for the experiments were in the form of pillars, deformed to successive compression strain of up to 35% respectively. After each strain ramp, monochromatic X-ray beam was used to measure the peak broadening.

2 Experimental Procedures

2.1 Materials

The single-crystal Cu/Nb nanoscale multilayers were grown by electron beam evaporation on single-crystal a-cut sapphire ($11\bar{2}0$) substrates (MTI Corporation, Richmond, CA) at elevated temperature of 200 °C. Deposition was

performed under a vacuum environment of at least 6.7×10^{-6} Pa at rates of 0.5 nm/s for Nb and 5 nm/s for Cu. Total thickness was about $3 \mu\text{m}$ and individual layer thickness of both Cu and Nb was 20 nm . The multilayer structure was formed by alternatively depositing layers of Nb and Cu as shown in Figure 1a. The first layer deposited was Nb, and it was deposited at the elevated temperature of 950°C for improved structural integrity with the substrate and the low deposition rate of 0.5 nm/s for improved orderly growth of the film as the seed layer for subsequent layers of both Cu and Nb. Further, the subsequent nanolayers were deposited at the temperature of 200°C with a deposition rate of 0.5 nm/s for Nb whereas a high deposition rate of 5 nm/s was used for Cu to achieve good quality and thermally stable (i.e., continuous layers) single-crystal multilayers. The growth of these nanoscale Cu/Nb single-crystal multilayers has been described thoroughly elsewhere [27]. Figure 1b shows XRD results of intensity vs. ϕ (ϕ) of both Cu and Nb nanolayer obtained by conventional laboratory XRD, which is described in Figure 1c. This intensity vs. ϕ (ϕ) - scans confirms the fabricated single crystal microstructure used in this study. A particular difference in the growth pattern compared to our earlier work [27] is, here, the nanolayer is epitaxially grown to a thickness of $\sim 3 \mu\text{m}$. Due to that, the individual single crystal nanolayers of both Cu and Nb have slight in-plane mis-orientation, which can be seen from the full width at half maximum (FWHM) of the XRD peaks (Figure 1b). The FWHM averaged across six peaks of both Cu and Nb is approximately $4 - 6^\circ$, which is higher than the earlier reported value of 0.09° [27]. The first Nb layer grew epitaxially and mostly $\{110\}$ textured on the single-crystal sapphire a-cut sapphire ($11\bar{2}0$)

substrate with only three in-plane variants (all $\{110\}$ textured but have different in-plane orientations, each shifted 60° from another). Subsequently, the Cu and Nb layers grew epitaxially and strongly $\{111\}$ and $\{110\}$ textured, respectively, on each other, and similarly with only two in-plane twin variants of Cu and the three in-plane variants mentioned above for Nb. The orientation of the multilayered film with respect to the substrate was found to be $\{110\}_{Nb} \parallel \{111\}_{Cu} \parallel \{11\bar{2}0\}_{Al_2O_3}$. The pillars were fabricated utilizing the annular milling method of focused ion beam (FIB) machining, where a Ga^+ ion beam is used to remove materials to shape a sample to high precision. The machined pillar has a nominal diameter (d_o) of $\sim 1.21 \mu m$ and a moderate vertical taper of $\sim 1.78^\circ$ as shown in Figure 1d. In the pillar of height $\sim 2.88 \mu m$, equal 20 nm thick layers of single crystal Cu and Nb are still present. The layer direction is perpendicular to the cylinder axis.

Fig. 1: Single crystal Cu/Nb multilayer, the diffraction scan and a pillar used in this study. a. TEM micrograph of the multilayer cross section, b. XRD results of Intensity vs. ϕ scan, c. X-ray conventions and d. typical FIB-milled pillar sample and its dimensions.

2.2 Methods

The uniaxial compression testing of these pillars was conducted at the Nano-Mechanical laboratory of Stanford University, using an Agilent/MTS Nanoin-denter XP (Santa Clara, CA) with a flat punch diamond tip. The nanoin-denter, which is a load-controlled instrument, was programmed to perform a nominally displacement- controlled test. In this method, the displacement

rate is calculated continuously during the compression test based on the measured displacement and time. When the measured displacement rate is below a specified value, the load is adjusted to maintain that particular displacement rate. This method is designed to simulate a constant displacement rate. Load-displacement data were collected in the continuous stiffness measurement mode of the instrument. The data obtained during compression were then converted to uniaxial true stresses and strains using the assumption that the plastic volume is conserved throughout this mostly homogeneous deformation.

The X-ray characterization were performed at the Advanced Light Source (ALS) of the Lawrence Berkeley National Laboratory, beamline 12.3.2 [32, 33]. The polychromatic X-Ray beam produced from a superconducting magnet source is refocused at the entrance of the experimental hutch by a 700 mm-long platinum-coated silicon toroidal mirror operating at a grazing angle of 4.5 mrad. Final focusing is done by Kirkpatrick-Baez (KB) focusing mirrors consisting of an orthogonal pair of 100 mm-long tungsten-coated silicon substrate bent to an elliptical shape. The Synchrotron X-ray micro-diffraction has nominally $0.8 \mu\text{m} \times 0.8 \mu\text{m}$ focused beam size but for monochromatic experiments as used in this study, an opening slit size of $2 \mu\text{m} \times 8 \mu\text{m}$ is more relevant in reference to the effective resolution, which still provides local microstructural and plasticity information of the sample [34]. The sample sits on a XY high precision stage (range of 10 cm), which is mounted on a coarse XYZ Huber stage (range of ± 15 mm in XY and Z). The diffraction patterns are collected with a DECTRIS Pilatus 1 M pixel area detector (active area of $179 \times 169 \text{ mm}^2$), which is placed at

a distance of approximately 140 mm from the sample. The CCD detector is comprised of pixels, from which the relative change in position and size of the peak is extracted. The resolution of this CCD detector used in the current study is $\sim 0.03^\circ$ [32]. The experimental setup in the beamline 12.3.2 provides the capabilities to obtain both kind of stress components *i.e. deviatoric and hydrostatic*. The technique at ALS Beamline 12.3.2 has previously been used to investigate plasticity of crystalline materials at sub-micron and nanometer scales[35–46]. Similar kind of experiments reported in the literature [47–53] applying synchrotron X-ray and neutron microdiffraction technique especially on Cu/Nb composites has been conducted in various beamlines around the world. More specifically the use of monochromatic beam to study and characterize microstructure has also been used and more completely described in this reference [34].

3 Results and Discussion

The mechanical response of the pillar samples deformed to varying compressive strain is presented. Then, the X-ray microdiffraction patterns from the same Cu/Nb nanolayered sample, before and after successive deformations are obtained using a monochromatic beam. The mechanical response and the microdiffraction observations of the nanolayer are correlated to confined layer slip, which is analyzed.

3.1 Mechanical response

In this study, three pillar samples (*each deformed with a different strain schedule up to 35%*) of similar dimensions were compressed using a flat punch. The measured load-displacement results of the samples shows variation within the nominal experimental scatter. Based on this affirmation, throughout this section, the results of only one pillar is discussed.

The true stress-strain mechanical response of the pillar sample from the compression tests are shown in Figure 2. The substrate effect is neglected due to no significant change in the results. The constant volume and homogeneous deformation model is used to interpret the load-displacement data. In this case, the volume measured from initial cross sectional area A_o (based on d_o) and length L_o of the undeformed pillar is equivalent to the final volume of the deformed sample, therefore: $A_o.L_o = A_P.L_P$, where A_P and L_P denotes final pillar area and length. To aid the understanding, at each strain ramp, the micrograph of the pillar with its topography taken after each cycle is shown in Table 1. It is seen from the stress-strain response (Figure 2a) that, pillar subjected to total true strain ε^T ($\varepsilon^{elastic} + \varepsilon^{plastic}$) of 1.6 and 3.3%, the single crystal Cu/Nb nanolayers deforms mostly linearly throughout the loading regime. The unloading of the pillar from a maximum strain of 1.6% results in a true plastic strain ($\varepsilon^{plastic}$) of $\sim 0.9\%$. Similarly, analyzing the unloading response of the Cu/Nb nanolayer subjected to a maximum strain of 3.3%, it can be clearly observed that it results in a true plastic strain ($\varepsilon^{plastic}$) of $\sim 2.5\%$. From the unloading curves, the composite elastic modulus obtained for ε^T of 1.6% and 3.3% is ~ 103.5 and ~ 105.8 *GPa* respec-

tively. The Young's modulus obtained from the experimental data reasonably agrees to the calculated value of $\sim 116.5 \text{ GPa}$ based on rule of mixtures. The true stress as seen from Figure 2a shows a peak value of $\sim 933 \text{ MPa}$ for 3.3% strained pillar as compared to sample with 1.6% which has a value of $\sim 845 \text{ MPa}$. This slight increase of maximum true stress at higher cumulative strain is obviously expected in mostly elastic regime. When the cumulative compressive strain increases to 12.8%, the nanolayers yield *i.e. onset of plastic deformation* at a stress of $\sim 1016 \text{ MPa}$ and thereafter shows a linear hardening behavior in the plastic zone until unloading with a peak stress of $\sim 1404 \text{ MPa}$. Tensile experimental response of a similar Cu/Nb free standing nanoscale multilayers with a $\{111\}_{Cu} \parallel \{110\}_{Nb}$ texture monitored through a synchrotron X-ray source confirms this clear elastic-plastic transition behavior [51]. From the unloading regime, the elastic modulus is seen to be $\sim 115.5 \text{ GPa}$, which agrees with the previous observed values. From the plastic flow curve, the modulus $H = \frac{d\sigma}{d\varepsilon^p}$, an equivalent measure of strain hardening rate, is found out to be $\sim 8.4 \text{ GPa}$. Further straining the pillar to 24.2% shows a much higher yield value of $\sim 1480 \text{ MPa}$ and an elastic modulus of $\sim 98 \text{ GPa}$, which is noted from the unloading curve. Moreover the nature of the plastic flow curve shows a considerable drop in strain hardening rate with a H value of $\sim 0.9 \text{ GPa}$ at a maximum strength of $\sim 1590 \text{ MPa}$ during unloading. These results of decrease in plastic strain hardening rate with respect to increase in compressive strain is consistent with experimental observations of Al/TiN multilayer composite [54]. Another interesting observation from this stress-strain curve is, elastic strain still being slightly present even during this large plastic deformation. To intuitively understand

this limit, one of the pillars was over-strained until 35% of plastic deformation, which showed near zero elastic strain in the nanoscale composite. The micrographs of the deformed pillar after each unloading as seen from Table 1 does not show any shear bands even at this large strains, while a bulge is noticed after a compressive strain of 12.8%. The elastic and plastic deformation behavior of the pillar obtained from these successive loading pattern confirms that, indeed the Cu/Nb nanolayer has a high flow strength before yield and is stable until plastic strain of 35%. After each loading-unloading, the X-ray microdiffraction experiments were conducted on the samples and its results quantitatively analyzed for dislocation evolution behavior, which will be discussed in the next section.

Fig. 2: True Stress vs. Strain of the pillar obtained from compression tests a. cumulative ε of 1.6 and 3.3%. and b. cumulative ε of 12.8 and 24.2%.

Tab. 1: Topography of the pillar after each strain ramp.

Total Strain ramp %	1.6	3.3	12.8	24.2
Deformed pillar

3.2 Dislocation evolution from X-ray microdiffraction analysis

The pillars were subjected to monochromatic beam of energy 10 keV after each strain ramp. We earlier explained in *Section 2.1* that, the individual nanolayers grown up to total film thickness of $\sim 3\mu m$ used in this study have in-plane mis-orientations which was further quantified using FWHM values. Due to this mis-orientation, we found that the Laue X-ray beam commonly used for these kind of plasticity analysis [13, 55] did not provide sufficient X-ray intensity to produce meaningful Laue reflections, that can shed light on the local plasticity and other microstructural features of the nanolayers, and this was the reason for using a monochromatic beam of energy 10 keV. The diffraction results shown in Figure 3 is obtained by transforming 2D synchrotron data using XMAS (a customized software developed at the Advanced Light Source, Beamline 12.3.2) software into the integrated intensity across various 2θ ($2\theta = \omega$ in laboratory XRD) angles. The complete details about the methodology which is slightly different compared to standard laboratory powder XRD, can be found in this reference [36]. The measured diffraction pattern of Cu and Nb is calibrated using a standard crystal structure of the same, to confirm the evidence of both these peaks. The highest peak intensity is observed at planes (311) (Figure 3a) and (310) (Figure 3b) for both Copper and Niobium respectively. Then, the peak broadening measured along χ direction at this intensity, of all the three pillars with respect to various compressive strain is analyzed using XMAS, to understand the formation of dislocation density. A schematic of peak broadening data

measured in this study is clearly explained in Figure 3c. The enlarged view of the peak (white square) shows a broadening which is estimated to be a measure of plasticity evolution and based on this, the further discussions follow.

Fig. 3: Calibrated diffraction pattern of a. Copper , b. Niobium and c. Quantitative data analysis of peak broadening for FWHM measurements subtracting the solid state cell effects.

The peak broadening of copper (311) and niobium (310) planes with increasing strains along χ is shown in Figure 4. The results show a common trend that, as the strain increase the peak broadening increase along χ direction for both copper (Figure 4a) and niobium (Figure 4b) nanolayers. Moreover, this increase in broadening of both copper and niobium single crystal nanolayers, is observed till a cumulative strain of $\sim 3.5\%$ and thereafter saturates till plastic strain of 35%. Along χ , the undeformed pillar samples show an initial peak broadening of $\sim 10.35 \pm 0.05^\circ$ in both Cu (311) and Nb (310) planes. Further investigating the copper nanolayer of all the samples in this study shows that, until a strain of 4%, peak broadening increases to $11.9 \pm 0.05^\circ$ along χ . Beyond the deformation regime of $\sim 3.5\%$, a marginal relative increase of $0.15 \pm 0.05^\circ$ is observed till 35% plastic strain. Similarly, the peak broadening of niobium layers also shows this trend. Till a cumulative compression strain of $\sim 3.5\%$, the peak broadening increases in χ to $12.0 \pm 0.1^\circ$, whereas further strains till 35% shows a relative increase of $0.1 \pm 0.1^\circ$.

Fig. 4: Peak broadening along χ with respect to various macroscopic compressive strains of a. Copper (311) and b. Niobium (310).

Peak broadening from the diffracted volume could provide information regarding plastic deformation [32, 35, 36, 34, 48]. From this, SSD density (ρ) as shown in Figure 5 were evaluated using a Cahn-Nye relation ($\rho = \frac{1}{R.b}$): R is obtained from local lattice curvature of the peak and b is the magnitude of Burgers vector. Even though, this technique is a very rough approximate, it provides an indication of quantifiable measure of dislocation density comparable with observations from the atomistic simulations [30] and TEM experiments [56]. It is important, however, to recognize that this model represents a methodology to obtain an estimate of dislocation density rather than its exact value. In addition, a few assumptions have to follow. First, parameters that can lead to peak broadening, such as, instrumentation, crystal size, and initial dislocation density due to the fabrication process, must remain identical for all samples. In this study all of these parameters are essentially the same for the pre- and postcompression XRD scans. Therefore, the only factor that affects the peak broadening is solely originated from dislocation activities in the nanolayers during the pillar compression. Second, considering the above other sources of broadening are rather ambiguous and thus their precise identification is mostly unavailable but that they are nominally the same for both states of the Cu/Nb pillar (before and after pillar compression), it is more meaningful here to just compare the relative values between them (i.e., the difference in the FWHMs between before and after the deformation) and simply assume all other broadening contribution to be at a certain fixed level (independent of the pillar deformation) which for convenience and simplicity here could just be taken as zero. By doing so, we can just apply the Cahn–Nye equation directly to the observed FWHMs

of the peaks between before and after the deformation. The SSD densities predicted using this approximation methodology based on the obtained values of FWHMs of the representative Cu and Nb peaks (as shown in Figures 4a and 4b) between before and after the deformation are all considered lower bound numbers.

The initial SSD (ρ) of undeformed copper (Figure 5a) and niobium (Figure 5b) nanolayers were $8.6E^{16}/m^2$ and $7.7E^{16}/m^2$ respectively. As the strain increases to $\sim 3.5\%$, SSD ρ also shows an increase to $10.0 \pm 0.05 E^{16}/m^2$ in copper and for niobium nanolayers, it increases to $9.0 \pm 0.05 E^{16}/m^2$. Further deformation till 35% of plastic strain in copper and niobium nanolayer shows a marginal relative increase of $0.1 \pm 0.05 E^{16}/m^2$ in ρ . The observations of peak broadening and SSD ρ at various mechanical deformation, shows evolution of dislocation in both the copper and niobium nanolayers.

From the macro-mechanical response of the composite, the dislocation behavior predicted has been confirmed with the microdiffraction observations. The increase of ρ up to strain of $\sim 3.5\%$ is consistent with the formation of parallel arrays of hairpin loops confined to individual layers. A plausible explanation for this kind of response from mechanical behavior is, Cu nanolayer alone deforms plastically whereas Nb behaves predominantly elastically [51]. The dislocation arrays (both parallel and oblique) formed due to the interaction of dislocations during the glide and some deposited at the interface, since the nanolayer has been subjected to plastic deformation, can cause significant work-hardening [17, 18]. There will also be a decrease in dislocation array distance λ as the plastic strains increase, which is related to the in-plane Burgers vector b through this relation: $\lambda = \frac{b}{\epsilon}$. Subsequently,

the stress field of the array which produces a force to repel a dislocation glide will eventually increase which will lead to higher flow stress as observed from Figure 2a. As the deformation increases, even at large plastic strains such as $\sim 35\%$, the composite exhibits high flow strength yet there is almost no increase in the SSD. Since the dislocations lie within the interface between the two individual nanolayer, during deformation they react with the other similarly deposited dislocations from the other nanolayer leading to crystal recovery/annihilations, which may be the cause for this behavior. The local stresses at the interfaces may have build to a level that dislocations can transmit across the interfaces to yield the Nb layer, which may be the reason for the lower work hardening rate, which can be observed from Figure 2b. The symmetric slip (balanced slip activities on multiple slip systems) lead to annihilations because of which the crystallographic stability of interfaces is maintained. It is also noted from these results that, in this small volume *i.e. nanolayers of single crystal Cu and Nb* the deformation does not multiply the dislocation leading to higher densities, which is not a phenomena observed in the bulk samples. This also confirms that, the Cu/Nb subjected to large plastic deformation is compatible at the nanoscale even though they have inherent differences of flow strength and hardening rate. These results from this study agree well with the direct experimental observations of confined layer slip reported by Li et al. [14] of this weak incoherent interfaces of nanoscale Cu/Nb multilayers.

Fig. 5: Statistically stored dislocation density with respect to various macroscopic compressive strains of a. Copper (3 1 1) and b. Niobium (3 1 0).

4 Conclusions

In this study, three pillars of nanoscale Cu/Nb multilayers were compressed using a flat diamond tip to a total strain ramp-up of 35%. After each deformation cycle, the samples were subjected to monochromatic X-ray beam of 10 keV at Advanced Light Source (ALS) of the Lawrence Berkeley National Laboratory, beamline 12.3.2. The peak broadening and the dislocation density of the Cu/Nb nanolayer with respect to various compressive strains is analyzed. The following conclusions from this study are as follows:

- Cu/Nb nanolayer pillars show all the expected behaviors under mechanical deformation/compression as have been reported widely in the literature. Both the Cu and Nb nanolayers were undergoing uniform compressive deformation to a large plastic strain exhibiting extreme deformability that has been attributed to crystal recovery mechanism. This model of deformation mechanism leading to crystal recovery has

been supported based on the synchrotron X-ray microdiffraction in the present study.

- The peak broadening of both Cu (3 1 1) and Nb (3 1 0) planes show an increase along χ direction with respect to various compressive strain, for all the pillars. A peak broadening of $\sim 10.35 \pm 0.05^\circ$ is noted in both undeformed copper and niobium nanolayers (Figures 4a and 4b) and the dislocation density quantified of the same shows $8.6 E^{16}/m^2$ and $7.7 E^{16}/m^2$ respectively (Figures 5a and 5b). Until successive compression strain of $\sim 3.5\%$, peak broadening increases in both copper ($11.9 \pm 0.05^\circ$) and niobium ($12.0 \pm 0.1^\circ$) nanolayers. Similarly the dislocation density also increases in both the copper ($10.0 \pm 0.05 E^{16}/m^2$) and niobium ($9.0 \pm 0.05 E^{16}/m^2$) nanolayers. The peak broadening and dislocation density of both copper and niobium nanolayer deformed from ~ 3.5 up to a strain of 35% shows negligible increase of $0.15 \pm 0.05^\circ$ and $0.1 \pm 0.05 E^{16}/m^2$ respectively. The peak broadening and dislocation density saturation after $\sim 3.5\%$ cumulative strain, confirms effect of crystal recovery/annihilations at the interface as proposed by confined layer slip theory.

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