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Berkovich nanoindentation study of 16 nm Cu/Nb ARB nanolaminate: effect of anisotropy on the surface pileup

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Abstract

Nanoindentation is widely used to investigate elastic modulus, hardness, and work hardening behaviour of nano- and micro-scale laminates. In this work, 16 nm accumulative roll bonding (ARB) Cu/Nb nanolaminate is used as a test material due to its interfacial anisotropy owing to the presence of contrasting interfaces along rolling (RD) and transverse direction (TD). Nanoindentation was performed along TD as well as RD of ARB Cu/Nb nanolaminate and then Scanning Probe Microscopy (SPM) data was collected to measure the pile-up along RD and TD in the Cu/Nb nanolaminate. 16 nm Cu/Nb ARB nanolaminate along RD was found to show higher surface pile-up than TD which is attributed to crystallographic and interfacial anisotropy resulting in (a) higher yield strength (low plasticity) along TD in comparison to RD and (b) high interfacial sliding in the case of TD resulting in less co-deformation of layers in comparison to RD. This work highlights the significant importance of the surface pile-up phenomenon in facilitating the study of anisotropic behavior in micro/nano-laminates.

keywords: nanoindentation, nanolayers, pile-up, plastic deformation, hardness, laminates

1 Introduction

Nanoindentation is widely used to measure the local elastic/plastic, and time-dependent mechanical properties of thin films and small volumes of bulk materials. Nevertheless, the stress state generated by indentation is complex as compared to typical tensile/compression experiments. Therefore, the deformation during indentation usually is inhomogeneous as compared to tensile/compression experiments [1–3]. Further, it is highly likely (in the case of nanolaminate) that indent will form within a single grain resulting in a highly anisotropic deformation [4–7]. This anisotropic deformation can give rise to plastic deformation-induced residual impression during indentation such as pile-up or sink-in characteristic of the nanolaminate. In fact, this crystallographic anisotropy dependence on interface-mediated plasticity in nanolaminate has recently been observed and studied in detail using advanced microstructural/nanomechanical characterization techniques [8–12]. These pileups (residual impressions) can also help to identify the anisotropy characteristic of the nanolaminate.

The test material used in the present analysis is 16 nm Cu/Nb nanolaminates fabricated through accumulative roll bonding (ARB). Cu/Nb ARB nanolaminates have become the subject of experimental and theoretical studies due to their outstanding strength, thermal stability and radiation resistance [13–15]. These unique properties make it a promising new generation material for a wide range of applications, such as novel metallic flexible/stretchable conductor technology. This particular sample (16 nm Cu/Nb nanolaminates), for instance, has been shown to exhibit significantly larger extent of interfacial sliding (localized shear region either on the interfaces or very near the interfaces) [8–10] compared to similar Cu/Nb nanolaminate samples with larger individual layer thickness [16]. Furthermore, within the same individual layer thickness, interfacial sliding has been shown to be substantially larger

in TD compared to in RD [8–10, 16]. Such a strong anisotropy could manifest in the surface pile-up (or sink-in) in a nanoindentation experiment. Early investigations into the mechanical properties of Cu/Nb nanolaminates relied exclusively on nanoindentation results [17, 18]. In general, varieties of dislocation-based models for multilayer material systems have been developed to fit flow strength data derived from the indentation measurements. However, due to the inhomogeneous stress state associated with indentation measurements, a better understanding of the plasticity due to the anisotropy associated with Cu/Nb nanolaminates is required. To this end, nanoindentation is performed with the major axis aligned along TD as well as RD to study their surface pile-up and then associate them to the variation in the plasticity observed along TD as well as RD.

In this study, the mechanical behavior of 16 nm Cu/Nb ARB nanolaminates was investigated through nanoindentation experiments. Later, Oliver-Pharr method was employed to obtain the hardness and reduced Young's modulus of the Cu/Nb nanolaminates. In the present work, we performed nanoindentation to obtain load-displacement curves and surface pileup along both RD and TD as a function of the indentation depth. Also, the trend for surface pileup for both RD and TD is analyzed and then associated with the variation in the plasticity along these directions.

2 Materials and Methods

2.1 Material

Multilayer Cu/Nb (copper/niobium) nanolaminates were prepared by accumulative roll bonding (ARB) in collaboration with Dr. Irene Beyerlein and Dr. Nathan Mara (both formerly at the Los Alamos National Laboratory). ARB samples were synthesized by repeated rolling of 1 mm Nb sheet sandwiched between two 0.5 mm Cu sheets. These Cu/Nb stacks were repeatedly cut into half and re-stacked and then re-rolled to obtain polycrystalline multilayer

Cu/Nb nanolaminate [19]. The rolling direction was kept constant throughout the ARB process. Cu/Nb ARB samples used for the analysis have 16 nm nominal individual layer thicknesses. A 4-axis diffractometer (XPert, PanAlytical), shown in Figure 1 of the supplementary data, equipped with a Cu anode ($\lambda = 1.54184 \text{ \AA}$) and operated at 40 kV and 30 mA was used for pole figure measurements as presented in Figures 1 and 2 for copper and niobium, respectively. Here, angle psi was varied from 0° to 75° whereas phi was varied from 0° to 360° (here, phi = 0° corresponds to RD and phi = 90° corresponds to TD) to obtain diffraction patterns from predominant crystallographic planes for both copper and niobium. Cu/Nb (16 nm) ARB interfaces consist of a mixture of the $\{112\} \langle 111 \rangle \text{ Cu} \parallel \{112\} \langle 110 \rangle \text{ Nb}$ and $\{552\} \langle 110 \rangle \text{ Cu} \parallel \{112\} \langle 110 \rangle \text{ Nb}$ [20]. Further details of the ARB process and the texture evolution can be found in the literature [13, 20]. The grains in these nanolaminates become extremely elongated, with aspect ratios exceeding 1:50:5 (ND:RD:TD) at layer thicknesses $\leq 65 \text{ nm}$ [21]. Further, plane strain deformation process of rolling generates anisotropy both in texture and grain morphology [22, 23]. Typically for Cu/Nb ARB samples, the stress required for the layers to shear along the RD is theoretically infinite as compared to 1.2 GPa for TD [24].

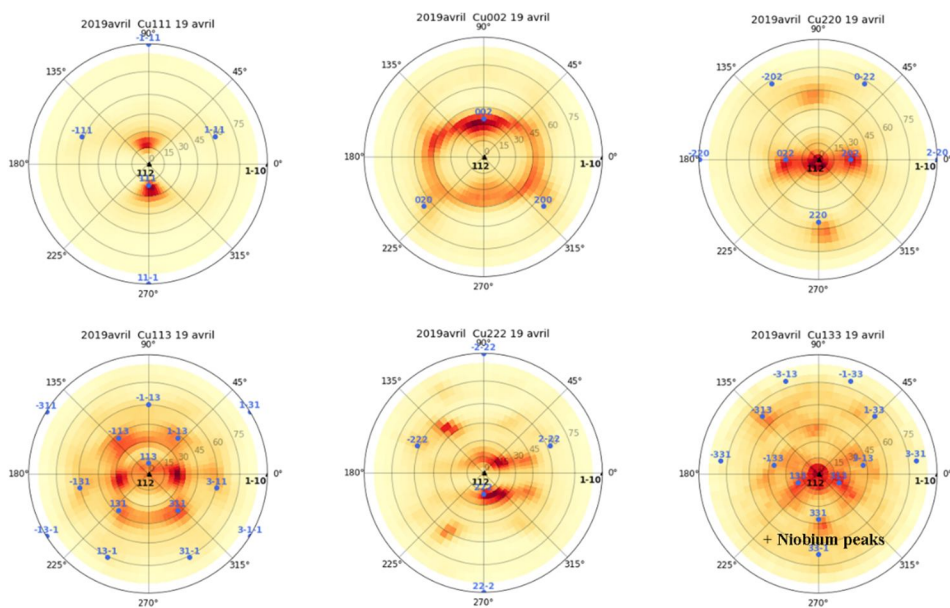


Figure 1. Pole Figure measurements of copper in Cu/Nb ARB nanolaminate using 4 axis diffractometer.

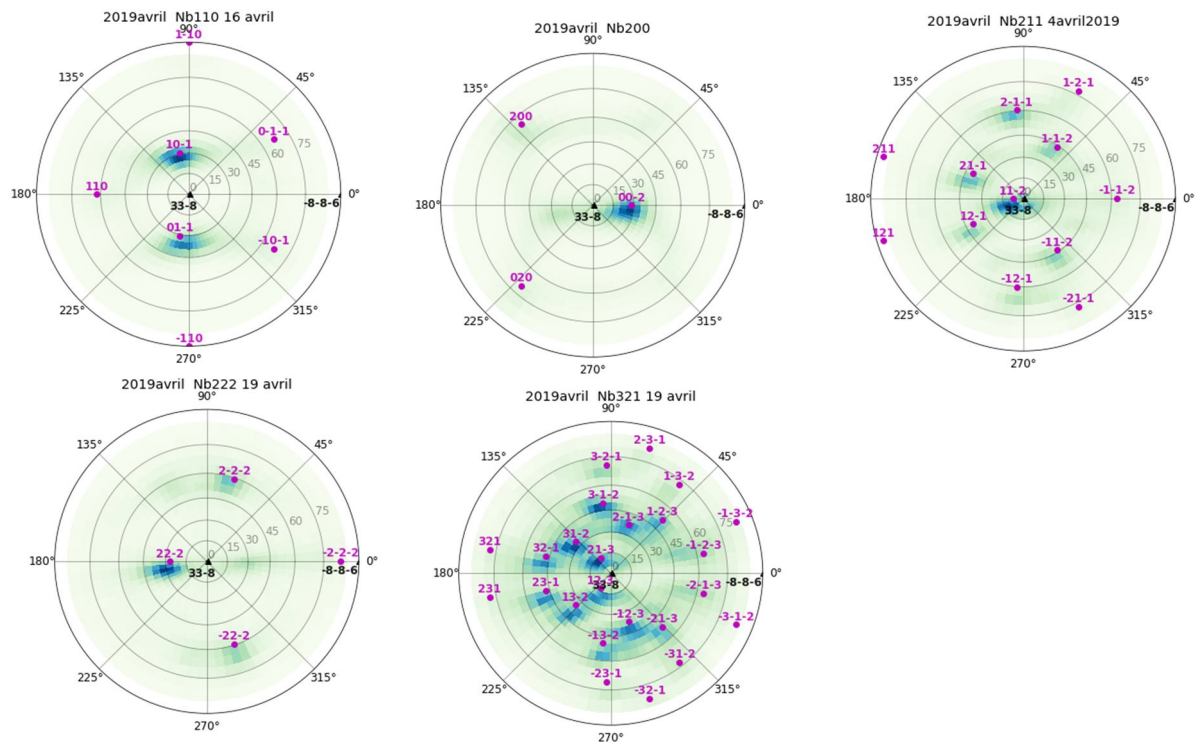


Figure 2. Pole Figure measurements of niobium in Cu/Nb ARB nanolaminate using 4 axis diffractometer.

2.2 Nanoindentation

Typically, during nanoindentation, a diamond indenter penetrates normally into a sample, and then the applied load (P) and displacement (h) are continuously recorded during the indentation cycle of loading and unloading. The load-displacement curve thus obtained is usually analyzed by the Oliver-Pharr method to obtain the mechanical properties of the samples. Nanoindentation experiments were performed using Hysitron Triboindenter TI 950 (make: Bruker Inc, USA) having displacement resolution ~ 0.2 nm and load resolution ~ 30 nN. The indentations were performed using a Berkovich tip in a continuous stiffness measurement mode. Prior to experiments, the Triboindenter is calibrated using a standard fused quartz sample for tip area function and machine compliance calibrations in order to precisely calculate contact stiffness using the Oliver-Pharr method. The tests were then performed in displacement control

mode with a loading displacement rate of 5 nm/s and an unloading displacement rate of 10 nm/s. The ratio of the specimen height to the maximum penetration depth is kept more than ~10 to avoid any substrate effect. The spacing between two indents was kept as ~20 μm to avoid any interaction between the plastic zone of indents.

2.3 Surface pileup profile

Scanning Probe Microscopy (SPM) included in Hysitron Triboindenter was used to map the profiles of the indents after nanoindentation. An area of $3 \times 3 \mu\text{m}^2$ including an indent was scanned to obtain profiles of the indents with high precision. Focused ion beam (FIB) milling (Zeiss Crossbeam 540) was used to polish the surface of the sample before indentation using low current of 1.5 nA at a voltage of 30 kV. The indented profiles were obtained on the polished sample surface to obtain height-profiles along RD and TD which were then used for measuring the pile-up along these directions.

3 Results and Discussion

3.1 Mechanical behaviour of 16 nm Cu/Nb ARB nanolaminate

Nanoindentation was used for the measurement of hardness, and reduced Young's modulus along RD and TD of 16 nm Cu/Nb ARB using Oliver-Pharr method [25]. Typically, in the case of Cu/Nb ARB nanolaminate, properties were found to vary with layer thickness. For instance, Beyerlein *et al.* [13] compared the hardness of Cu/Nb ARB nanolaminate as a function of layer thickness ranging from 714 nm to 7 nm. The hardness increased with the reduction in the layer thickness to an ultrahigh value of 5 GPa for layer thickness equal to 7 nm. This value is two orders of magnitude higher than those of Cu or Nb bulk materials.

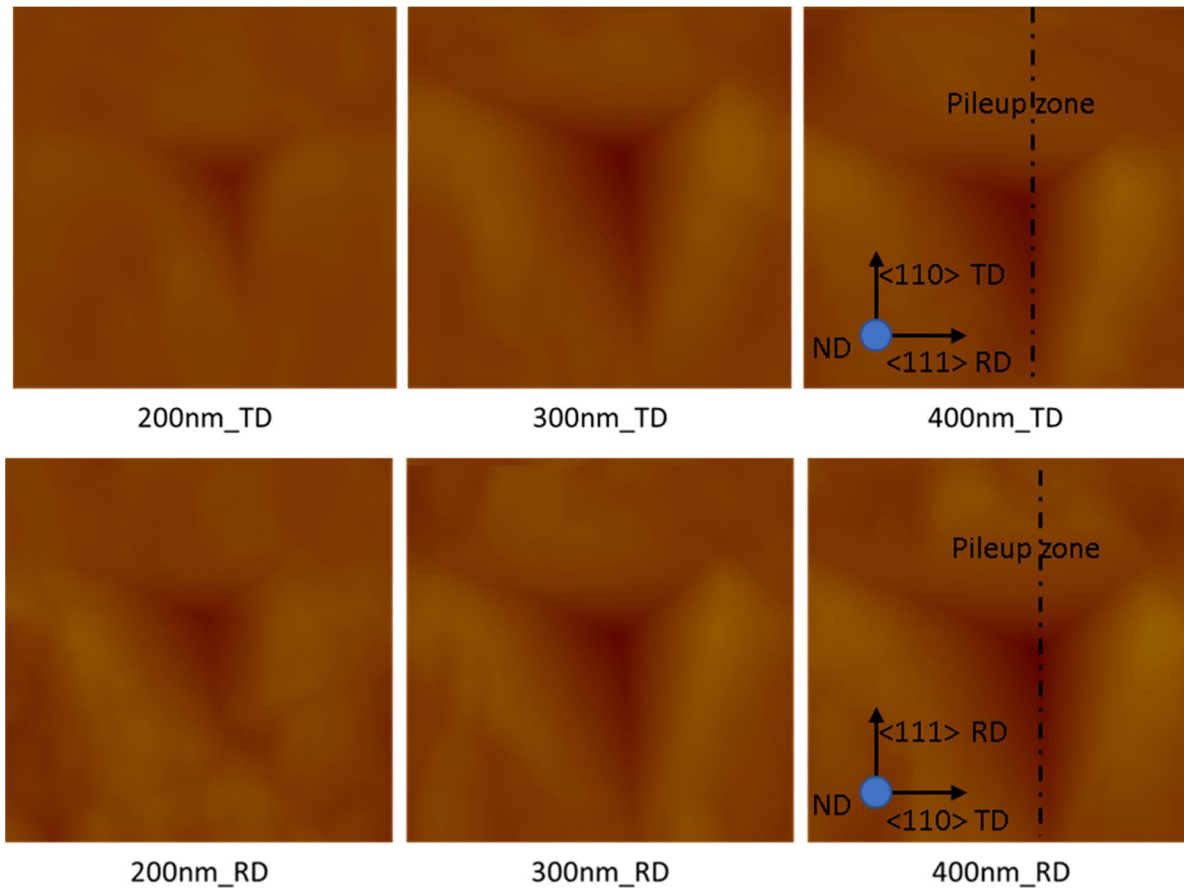


Figure 3. Figure shows Scanning Probe Microscopy (SPM) images of the indents for TD and RD as a function of indentation depth. The scanned area was $3 \times 3 \mu\text{m}^2$. These images are obtained after each indentation to accurately measure the height-profiles and corresponding pileups. Crystallographic directions are indicated for Niobium. The height profile is obtained along the major axis shown with a dotted line.

In this analysis, hardness and reduced Young's modulus of 16 nm Cu/Nb ARB nanolaminate were calculated as a function of indentation depth. Representative indents as a function of indentation depth are shown for TD and RD in Figure 3, and their corresponding load-displacement curves are shown in Figure 4. The experimentally obtained nanoindentation load-displacement curves depict an increase in the applied load with an increase in the indentation depth for both TD and RD. For both TD and RD, the indentation depth applied were 200 nm, 300 nm and 400 nm. Nevertheless, in the case of TD, the maximum indentation depth reached is 356.95 in comparison to applied 400 nm in the case of RD. The low achievable indentation

depth in the case of TD is attributed to its high yield in comparison to RD [26] which is also responsible for low pileup for TD in comparison to RD. Later, Oliver-Pharr method is used to calculate hardness and reduced Young's modulus from experimentally obtained load-displacement curves. The hardness of 16 nm Cu/Nb ARB was found to reduce from 4.33 GPa to 2.72 GPa and the Young's modulus reduced from 107.24 GPa to 92.72 GPa with an increase in the indentation depth from 200 nm to 400 nm.

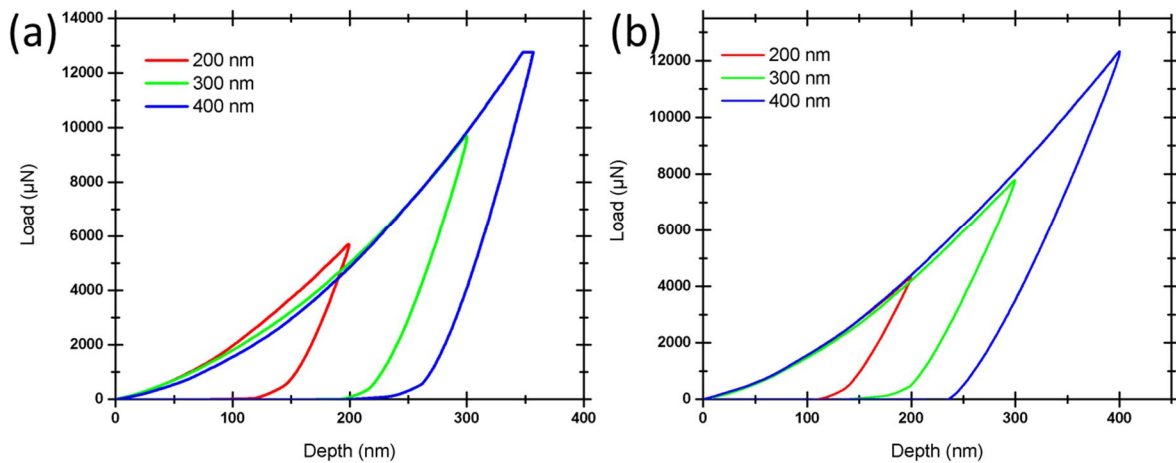


Figure 4. Figure shows the typical load-displacement curves obtained through nanoindentation for (a) TD and (b) RD as a function of indentation depth.

3.2 Pileup as a function of anisotropy of the 16 nm Cu/Nb ARB nanolaminate

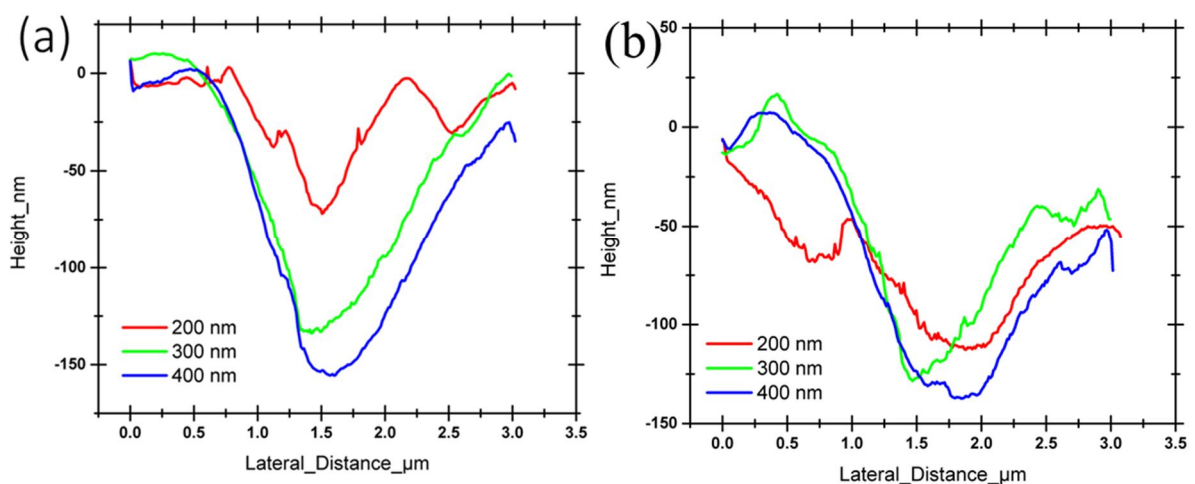


Figure 5. Height profiles along the major axis of indents used for measuring pileups along (a) TD and (b) RD as a function of the indentation depth.

The profiles of the indents were scanned using SPM. Height-profiles curves along the major axis of the indents were obtained which assisted in measuring the pileup along TD and RD. Figure 5 shows height-profile curves along TD and RD as a function of the indentation depth ranging from 200 nm to 400 nm. For the indentation depth less than 200 nm (such as 50 nm, 100 nm), pileups were small and, in some cases, comparable to the surface roughness and so were not easily distinguishable by SPM. As can be seen from Figure 6, the pile-up heights increase with an increase in the indentation depth for both RD and TD. Comparison of the pile-up heights along RD and TD show an increase in the pile-up height for RD in comparison to TD with an increase in the indentation depth.

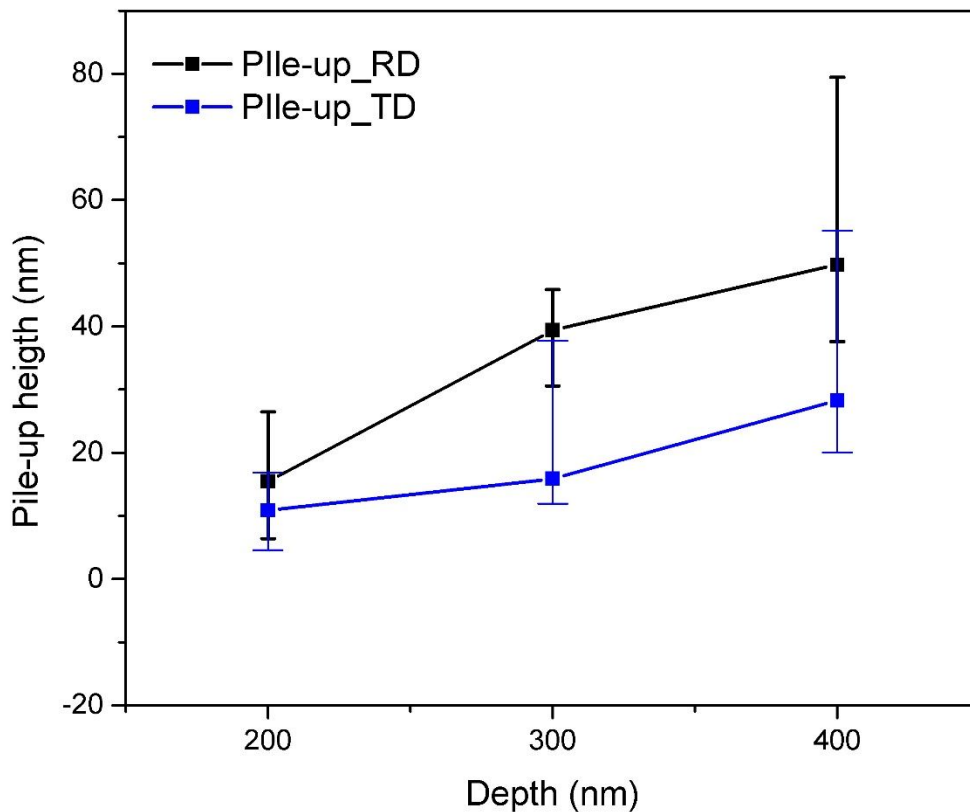


Figure 6. Comparison of the pile-up heights along RD and TD Cu/Nb nanolaminate as a function of the indentation depth.

4 Discussion

4.1 Nanoindentation

During nanoindentation tests for nanolaminates, the dislocation nucleation, and their propagation is complex and not well understood. Soft, easily hardened materials sink-in whereas harder, work-hardened materials pileup [27]. In general, dislocations will be generated in the material near the tip of the indenter. The propagation of these dislocations along the available slip planes in the material will be governed by the applied load (i.e. resolved shear stress along the slip plane). If resolved shear stress is higher than the critical resolved shear stress (CRSS) required for the propagation of the dislocations, these dislocations could propagate along the slip plane and could move downwards into the material or cross-slip to the surface of the indent generating pileup. In case of heavily work-hardened (low yield stress) RD (due to repetitive rolling during fabrication), RD's capacity for downward dislocation motion into the indent could reach its peak sooner than in TD, thus increasing the propensity for cross-slip on the available slip planes and then generating a higher pileup in RD in comparison to TD.

4.2 Pileup profile

The pile-up of material during nanoindentation is highly dependent on its crystallographic texture. Further, the height of the pileup can vary on the same surface plane indicating a strong influence of slip geometry leading to plastic anisotropy as observed along RD and TD in 16 nm Cu/Nb ARB nanolaminate. In the case of Cu/Nb ARB nanolaminate, the crystallographic axes aligned along the normal direction (ND) can range from $[4\ 4\ 11]$, $[110]$ to $[112]$ depending on the Cu layer thickness [13]. Similarly, for Nb, the ND can range from $[3\ 3\ -8]$, to $[112]$ depending on the layer thickness. For the 16 nm Cu/Nb ARB sample, 4 axis diffractometer was used to obtain crystallographic axes aligned along the normal direction, rolling direction and

transverse direction (see Figures 1 and 2). For Cu, ND is [1 1 2], RD is [1 -1 0] and TD is [-1 -1 1] and for Nb, ND is [1 1 2], RD is [1 1 1] and TD is [1 1 0]. Typically, in order to document the effect of crystallographic texture on the nature of surface pileup, it is advisable to document activated slip systems with respect to TD and RD [28]. In particular, for FCC Cu, there are 12 slip systems. Dislocation along a slip plane is triggered when the shear stress along that slip plane reaches the critical resolved shear stress. Therefore, CRSS (or Schmid factor) can be calculated for available slip systems along RD and TD to document activated slip systems along RD and TD. Nevertheless, Berkovich indenter (in comparison to conical or spherical indenter) imposes a complex stress field under the indenter which can result in non-uniformly activated slip systems which make it difficult to directly compare Schmid factor calculations to the experimental results such as surface pileups.

Apart from the material plasticity, plasticity at the Cu/Nb interface also affects the pileup. Demkowicz *et al.* [24] had documented interface shear strength along TD is 1.2 GPa whereas it is infinite along RD. The infinite shear strength along RD allows for co-deformation of layers and subsequently high pileup in comparison to TD where the interface has the capacity of slide [8–12, 16].

5 Conclusions

The present analysis performed nanoindentation on 16 nm Cu/Nb ARB nanolaminates as a function of the indentation depth. The surface pileup was found to be higher along RD than along TD which is attributed to their crystallographic and interfacial anisotropy. These anisotropy along RD and TD results in higher yield strength (low plasticity) along TD in comparison to RD. Interfacial anisotropy also contributes to interfacial sliding in the case of TD resulting in less co-deformation of layers in comparison to RD. Both these factors contribute to higher pileup along RD in comparison to TD.

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