LA-UR- 98-2979

CONF-980405-

Title: Plastic Behaviour of Cu/Ni Multilayers

Author(s): M. Verdier, LANL
M. Niewczas, McMaster Univ.
J.D. Embury, LANL
M. Hawley, LANL
M. Nastasi, LANL
H. Kung, LANL

Submitted to: MRS Spring Meeting
San Francisco, CA
April 13, 19998

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Plastic Behaviour of Cu / Ni Multilayers

M. Verdier⁽¹⁾, M. Niewczas⁽²⁾, J.D. Embury^(1,2), M.Hawley⁽¹⁾, M.Nastasi⁽¹⁾, H.Kung⁽¹⁾

(1) Los Alamos Nat Lab., CMS, MS K765, Los Alamos, NM 87545.

(2) McMaster University, Dept of Materials Science and Engineering, Hamilton, ON, Canada.

ABSTRACT

In order to study the plasticity in Cu-Ni multilayers deposited on single crystals of Cu and etched Si, we have investigated the mechanical properties of the multilayers both by nanoindentation measurements, and by the transmission of well characterized dislocations from the underlying substrate by tensile deformation of Cu single crystals.

Various multilayers were deposited by physical vapor deposition with layer thicknesses varying between 1000 and 20 Angstroms (for a total thickness between 0.8 and 1 μm). Two types of experiments were designed. The first one aimed at injecting, in a controlled way, some dislocations from the substrate into the multilayers; the second type of experiment concerned the structure of the multilayer surface after having plastically pushed the material away from a nanoindenter.

This communication reports the results from the nanoindentation measurements, as well as the observations of slip on the surface. We observed through the injection of dislocations by nanoindentation that the multilayers increase in strength with refinement of the layer structure but at thicknesses below 35Å exhibits a softening behaviour. Also observation of the upheaval around the nanoindent showed an evolution from slip lines to more spread plasticity with refinement of the layer structure.

INTRODUCTION

The mechanical properties of nanocrystalline materials have been the subject of numerous experimental and theoretical studies in recent years [1,2]. Among the various processing methods available for production of nanoscale materials, Physical Vapor Deposition of metallic multilayers is ideal for the study of both the influence of the scale of the structure and the effect of interface on the mechanical properties. For these fine scale composite materials, the yield strength increases with decreasing layer thickness, and may reach values close to the theoretical strength (a small fraction of the Young modulus). Several models attempt to describe the evolution of the yield strength of the composite with layer thickness, such as the Hall-Petch model for dislocation pile-up at the interface, or the behaviour of a single dislocation in a constrained layer [3]. Several review articles detail these approaches [4-6]. There is, however, no general model able to describe the evolution of the yield strength over a large scale range (from 1 micron down to 2 nm layer thickness), and questions such as the relevance of the concept of dislocations as used for the bulk material behaviour at the nanometer scale, still exist.

Moreover, experimental observations of the characteristics of deformation of multilayers are scarce. One reason is probably linked to the difficulty in controlling deformation in such structures when they are a micron in thickness.

In the present work, we report the observations of deformed surfaces of Cu/Ni multilayers in relationship with their strength measured by nanoindentation. Two types of experiments were carried out to induce plasticity in these multilayers. The first one aimed at injecting in a controlled way some dislocations from the substrate into the multilayers; the second type of experiment concerned the surface structure of the multilayer after having plastically pushed material away from a nanoindenter :

- The Cu single crystal used as substrates had an crystallographic orientation allowing the activation of only one dislocation system during tensile tests at liquid nitrogen temperature. Thus with this experimental procedure, we can control the transmission of dislocations through the

multilayer and study the influence of layer thickness on this process (fig.1a). After tensile testing of the substrate, the slip patterns at the surface of the multilayer evolved with layer thicknesses from slip lines to fracture lines, still oriented along the substrate slip lines.

- The nanoindenter was used to penetrate about 80% thickness of the multilayer deposited on Si substrates (fig. 1b). The resulting patterns of upheaval of material around the indent was characterized by AFM.

Figure 1a. Injection of dislocation through the multilayers interface.

Figure 1b. Plastic flow of the multilayer generated by nanoindentation (upheaval)

EXPERIMENTAL

The Cu/Ni system was chosen for these experiments because the bulk plasticity behaviour for these metals is well known. The misfit in the $[100]$ interface is 2.8%, and the ratio of their shear modulus G is $G_{Ni}=1.6 G_{Cu}$.

Cu/Ni multilayers were deposited by e-beam evaporation on the (321) plane of Cu single crystal, grown from high purity Cu using the Bridgman technique (see fig. 2) and on the $\{100\}$ Si surface. The total thickness of the multilayers was $1 \mu\text{m}$ on the Cu substrates and around $0.8 \mu\text{m}$ on Si. The Cu single crystal were electropolished prior to introduction in the e-beam chamber then coated with a Cu seed layer (100nm) deposited at 350°C . The Cu/Ni layers were grown at room temperature because of the total miscibility of this system at elevated temperature. The Si substrates were cleaned by etching with a 10% HF solution prior to deposition of a Cu seed layer (100 nm) at room temperature [7]). The deposition rate was $5 \text{ \AA/s}$ for all layers at a the vacuum around 5×10^{-8} Torr.

The hardness and indentation modulus of the multilayers were measured using a indentation load-depth sensing apparatus (Nano Indenter® II). A continuous stiffness technique was used for nano-indentation at a displacement rate of 2 nm/s with a Berkovich tip.

The geometry used for the tensile test of the multilayer on the Cu single crystal substrates is shown in figure 2.

The Cu crystals ($0.3 \times 0.3 \times 3 \text{ cm}$) were grown along a $[541]$ direction which has a square cross-section ($\{321\}$ and $\{111\}$ planes). This geometry allows the activation of one slip system (here the slip plane is on the $(11\bar{1})$ plane with b along $[101]$) when tensile stress is applied along the $[541]$ direction. To extend this stage I of work hardening, tensile tests were carried at 77K up to $\epsilon \approx 2.5\%$ at $d\epsilon/dt \approx 10^{-4} \text{ s}^{-1}$. Multilayers were deposited on the $\{321\}$ plane of the crystals, so that only edge character dislocation coming from the substrate would penetrate the interfaces. The traces of $(11\bar{1})$ planes on the $(12\bar{3})$ plane make an angle of 64.9° with the edge $[541]$ of the crystal ; they are 24.1° off the normal of the tensile stress axis on the deposition plane.

Figure 2. Geometry of the multilayer+Cu substrate

The multilayers deposited on {100} surface of etched Si substrates were indented down to 600 nm (80% of the total thickness).

Observations of the surface of the deformed multilayers by tensile tests were done with a standard optical microscope with Nomarsky contrast and by SEM. The nanoindents on the Si substrates were imaged simultaneously with atomic force microscope (AFM) in height mode (tapping mode), in lift force gradient mode and magnetic force microscopy both with using a Cr coated tip using a Nanoscope III (Digital Instr.) AFM.

RESULTS and DISCUSSION

A- Mechanical properties measured by nanoindentation -

The evolution of hardness (H) and apparent Young's modulus (E) of the films versus the layer thickness λ are shown in figure 3a and 3b, together with other Cu/Metal systems studied at Los Alamos National Lab. Figure 3a is a Hall-Petch type plot (H-P) showing that an invariant law relating hardness with $1/\sqrt{\lambda}$ does not exist on the whole range of λ . The initial slope of the H-P plot can be related to the shear modulus of the harder phase [8] and can be associated with a pile up mechanism of dislocation at the interfaces of each layer. Under a critical layer thickness (λ between 500 and 200 Å), the H-P slope decreases and, in the case of Cu/Ni exhibits an inverse Hall Petch behaviour. This softening effect has been reported in different systems [9] and observed in tensile tests of free standing Cu/Ni multilayers [10,11], and in numerical simulation of deformation of nanograins of Cu [2].

Fig. 3a : Hall-Petch plot of hardness of Cu/X systems

Fig.3b : Evolution of E versus layer thickness

The effect of indenting the multilayers on a softer substrate does not modify this general behaviour as can be seen in fig.3a for the Cu/Ni deposited on the Cu substrates or NaCl single crystals.

Concerning the Young modulus of the multilayers, although no super-modulus effect was observed, a small increase of 10% was observed with decreasing layer thickness between 500 Å and 50 Å. This increase is interpreted as an effect of the intrinsic tensile stresses developed during deposition which vary with the layer thicknesses [12].

B- Injection of $[101](11\bar{1})$ dislocations : tensile tests on Cu [541] substrates -

The tensile curves (τ, γ) of the system multilayer + Cu single crystal substrate are shown in fig. 4. The critical resolved shear stress (CRSS) for the single crystal of around 1.5 MPa is shown with a dashed line. All curves exhibit a linear stress-strain relation characteristic of the stage I of work hardening. The presence of a multilayer on one surface of the crystal has modified the CRSS of the system : for λ in the 1000-500 Å range, we observed a net increase of the CRSS, while for $\lambda < 500$ Å, the measured CRSS was close to the Cu single crystal without any multilayer.

We have interpreted this effect in relationship with the observations of the deformed multilayer after the test.

Fig. 4 : Stress-strain curves of Cu/Ni multilayers deposited on Cu single crystal

It appears that for large layer thicknesses ($\lambda=1000$ and 500 Å), slip bands are observed on the surface of the multilayer, fig. 5a. The slip bands observed on the $\lambda=500$ Å multilayer are however less sharp than the one observed for $\lambda=1000$ Å.

The orientation of the bands correspond to the traces of the activated $(11\bar{1})$ slip plane, and are indicated with a white arrow. This slip transmission confirms the same crystallographic orientation between the substrate and the multilayer.

Fig. 5a Slip bands on the surface of Cu/Ni multilayer, $\lambda=1000$ Å oriented along the traces of $(11\bar{1})$ planes .

Fig. 5b Cracks on the surface of Cu/Ni multilayer, $\lambda=200$ Å. The cracks are oriented along the traces of $(11\bar{1})$ slip planes.

For all the deformed multilayers with layer thicknesses less than 500 Å, the surfaces exhibited some fine straight fracture cracks orientated along the traces of the $(11\bar{1})$ slip planes, with a spatial distribution equivalent to that observed for the slip bands.

Fracture along the slip plane direction, rather than normal to the applied tensile stress indicates that the interface in those films did not transmit the dislocations coming from the Cu substrate, fig. 5b. Instead the films relaxed by fracture. One possible explanation is that the increasing intrinsic stress (due to evaporation) linked with the decreasing layer size plays an important role to reach the critical toughness of the film, triggered by the pile-up of few edge dislocation emanating from the Cu substrate.

In the initial stage of tensile deformation, the multilayers ($\lambda < 500$ Å) appear to crack, resulting in no hardening effect on the CRSS.

B- Surface aspect of upheaval of nanoindented multilayers -

The second set of experiments concerns the observation of the surface of plastically deformed multilayers. We have observed by AFM techniques plastic flow in the upheaval of material around nanoindents. The depth of the indents were at 80% of the total film thickness deposited on etched Si substrates. The upheaval was between 150 to 200 nm high over a micron length. The height, pseudo-derivative height, and force gradient of the magnetic polarization were

imaged. The height derivative helps to visualize the presence of steps or slip bands at the surface of the sample, while the vertical magnetic polarization (induced by the Ni coupling between the layers [13]) can indicate some buried cracks in the structure.

Figure 6a shows a well defined squared array of slip lines around the indent on the $\lambda=500 \text{ \AA}$ multilayer. The square shape corresponds to the traces of (111) slip planes of the Cu/Ni multilayer projected on the (100) surface. This indicates that a dislocation based mechanism still exist for layer thicknesses greater than $500 \text{ \AA}$. The vertical magnetic polarization of the indent, fig. 6b shows the pattern of the domains around the indent. The interpretation of the signal around and in the indent is difficult due to the effect of elastic strain on the magnetic domain structure ([13]), but the presence of black straight lines in the indent could indicate the presence of cracks in the multilayer or more likely in the Si substrate.

Fig. 6a : height derivative, $\lambda=500 \text{ \AA}$

Fig. 6b : vertical magnetic polarization

For smaller layer thicknesses, evidence of dislocation slip lines around the indent are less obvious : the multilayer seems to deform more homogenously on the upheaval as it can be seen in fig. 7a for $\lambda=25 \text{ \AA}$.

Fig. 7a : height derivative, $\lambda=25 \text{ \AA}$

Fig. 7b : vertical magnetic polarization

Figure 7b indicates a coarser structure for the magnetic domain due to a better coupling between the layers. One can note that the effect of strain on the magnetic polarization does not extend further than the plastic deformation linked with the plastic flow of material.

CONCLUSION

The evolution of hardness versus the layer thickness of the nanocomposite Cu/Ni shows that there is not a unique correlation between the mechanical properties and the scale of the layers. Several parameters can change while changing the scale of the structure :

- the evolution of the internal stress, which will have an effect on the core properties of the dislocations, and as well on the onset of the plastic deformation,
- the difference in slip transmission with the different nature of the interfaces; this effect will create a competition between plastic relaxation and propagation of instabilities,
- the difference in slip distribution, which will more or less influence the homogenous character of the plastic flow.

In this work, we were able to see that the slip distribution is finer with smaller layer thickness (λ) from the AFM study of indented multilayers. Moreover, a different permeability of the interfaces with dislocation pile-up was evidenced from the tensile experiments on Cu single crystal substrates.

These type of experiments are currently developed in an effort to measure the internal stress of the structures and to perform a detailed basis for the structural characterization of deformed multilayers by TEM and AFM in-situ observation during the compression of Cu single crystal substrates.

ACKNOWLEDGMENTS

The authors would like to thank Dr. Basinski for the elaboration of Cu single crystals, J.P. Hirth for fruitful discussion, and G.W. Brown for his help with the AFM. This work was supported by an LDRD program (XAEM) at Los Alamos National Laboratory (D.O.E).

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