IN SITU TEM MONITORING OF DISLOCATION MECHANISMS IN THIN METALLIC FILMS

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ABSTRACT

The plastic deformation of thin metallic films has been studied by mean of in-situ Transmission Electron Microscopy (in-situ TEM). In the case of films deposited on a rigid substrate, thermal stresses are used to plastically strain the film. Strain is directly related to the difference in coefficient of linear expansion between the film and the underlying substrate and does not rise above a couple % per thermal cycle. We performed similar tests inside a TEM, on plane view and cross sectional specimens. This allows for direct and real time observation of most dislocation based mechanisms, including their interaction with interfaces, grain boundaries and free surfaces. It has been shown that dislocation-based strain hardening never occurs among successive thermal cycles. This holds true for polycrystalline films deposited on an amorphous diffusion barrier or for pseudo epitaxial single crystalline film on sapphire substrate. The nature of the interface emerges as the key point in all these experiments. We will try to emphasize the role of these interfaces, which is strongly related to their nature, i.e. amorphous/crystalline or crystalline/crystalline. It is interesting to note that, sometimes at variance to what dislocation theory predicts, rigid substrates can act as dislocation sinks.

1. INTRODUCTION

Understanding the plastic deformation of thin metallic films deposited on rigid substrates bear some interest both from industrial and theoretical points of view.

Thermal stresses are one of the potential cause of failure in microprocessors as they can lead to large permanent deformation of the metallic interconnects. The amount of total strain in the metal layers is related to both the coefficient of linear expansion mismatch between the different materials and to the number and amplitude of the imposed thermal cycles. In smaller lines, the issue of plastic deformation becomes actually less important since the ratio of plastic/elastic strain usually decreases with interconnect dimensions.

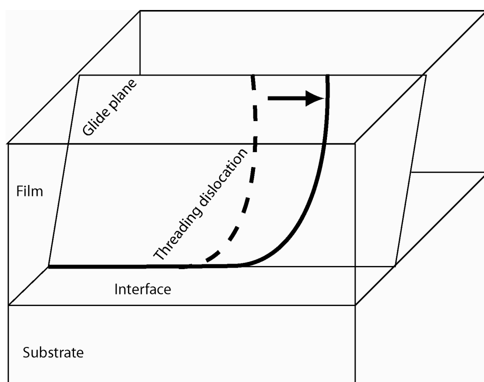


Figure 1: A threading dislocation moving in a thin film lays down an interfacial segment

On a theoretical point of view, the basic phenomena of plasticity in thin metallic films remain largely uncovered. In this work, we will consider mainly the thermo-plastic behavior of metallic films bounded to a substrate. Free standing films will only be brought in for comparison purposes since data concerning their stress/strain exhibit significant scatter. On the other hand, substrate-supported films can be accurately characterized through wafer-curvature laser measurements coupled with the Stoney formula [1]. The process of depositing metallic films with a known microstructure onto

silicon substrates is also well controlled. One of the drawbacks of the wafer-curvature technique is that one has to vary the temperature to have a stress evolution and thus some eventual plastic deformation if the difference between the coefficient of linear expansion of both film and substrate is sufficient. The yield stress of the metallic film also usually depends on the temperature, the thermal history of the film, and as it was early shown, on the film thickness [2]. If d is the film thickness, the yield stress was found to vary as 1/d. This result was readily interpreted as the result of threading dislocations channeling through the film : the thinner the film, the smaller the radius of curvature of mobile dislocations and thus higher the stress needed to make them move [3], [4], [5]. This model assumes that, as in the earlier Matthews-Blakeslee model [6] that describes relaxation processes in epitaxial semiconductor layers, an interfacial dislocation is laid down on the metal side of the metal/substrate interface (Fig. 1).

Another approach consisted in combining the Frost and Ashby deformation maps [7] with temperature and strain data provided by wafer curvature experiments. From these maps, established for bulk materials, it is theoretically possible to calculate a strain rate for a given stress and temperature and thus determining which mechanism will prevail, based on the fastest rate. This way of investigation supposes that deformation mechanisms are similar in bulk and thin film materials and that they operate for the same stress and temperature ranges.

TEM and in-situ investigations of the microstructural changes in deposited films have started as early as 1969 [8], [9], showing dislocation activity or grain growth. Since then, multiple attempts to dynamically observe what was occurring within the film have been made, focusing more on the link between microstructural observations and macroscopic behaviour [10], [11], [12], with sometimes conflicting results. Pursuing efforts lead to two key experimental observations: the disappearance of interfacial dislocations in an Al film deposited on Si [13] and the occurrence of parallel glide dislocations in ultrathin Cu films [14]. While the instability of interfacial dislocations has been confirmed by cross-sectional in situ TEM studies [15], parallel glide has been interpreted as the result of grain boundary diffusion [16]. Because they are not expected by classical dislocation theory, these mechanisms demonstrate that current models need to be implemented or renewed. Additional and more systematic in-situ TEM observations are currently performed, aiming to a more comprehensive understanding of thin metallic film plasticity and addressing current open questions such as strain hardening, dislocation interaction with interfaces and nucleation, proportion of diffusive mechanisms in stress relaxation.

2. EXPERIMENTAL

The main difficulty with in-situ TEM is to work with samples that have a thickness about or inferior to 300 nm. Not only the stress state is different compared to the initial film over substrate configuration, but additional thin foil effects and potential artifacts such as electron

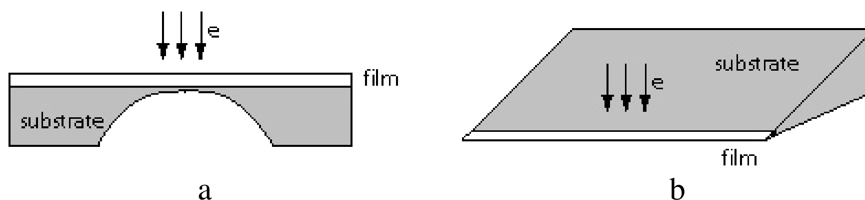


Figure 2: Possible configurations for in-situ TEM testing : a) plane view, b) cross-section. Arrows indicate the direction of the electron beam.

irradiation have to be considered. To reproduce the thermal stresses felt by a metallic film attached to a substrate, two configurations are available: plane view (Fig. 2a) or cross-section (Fig. 2b).

The plane view configuration is only available for film thicknesses that are below 300 nm in the better cases (light metals). In these favorable cases, the observable window may include a small amount of the remaining substrate, but this later one will not provide an equivalent amount of stress and strain as compared to the complete substrate. In the cross-section configuration, FEM calculations lead to an estimated stress level of about 70% of the "bulk" sample stress, but mainly unidirectional. Samples are generally prepared using a dimpler (plane-view configuration) or tripod (cross-section), while final polishing is obtained through low angle ion beam polishing. The resulting samples are glued to a copper grid using a heat resistant epoxy or a water-based cement. As it will be shown here, early experiments using a high voltage TEM led to irradiation of some films, providing unexpected dislocation sources. Subsequently, in-situ experiments were carried out on 200 keV TEMs equipped with double-tilt heating or straining stages and video-rate CCD cameras. Temperature excursions range between room temperature and 450°C to 600°C (depending on the nature of the film). In order to validate the in-situ observations, TEM investigations were also run on full size samples that were thermally cycled outside the TEM ("post-mortem" TEM). Investigated samples include Au, Al and alloyed Al polycrystalline films deposited on diffusion barrier coated Si wafers and pseudo single grain Cu films deposited onto sapphire substrates. Thicknesses range from 100 to 1000 nm. The purpose of using different substrates is to investigate different type of interfaces : crystal/amorphous in the case of Au and Al and crystal/crystal in the case of Cu.

3. RESULTS

3.1 Grain growth and microstructure evolution.

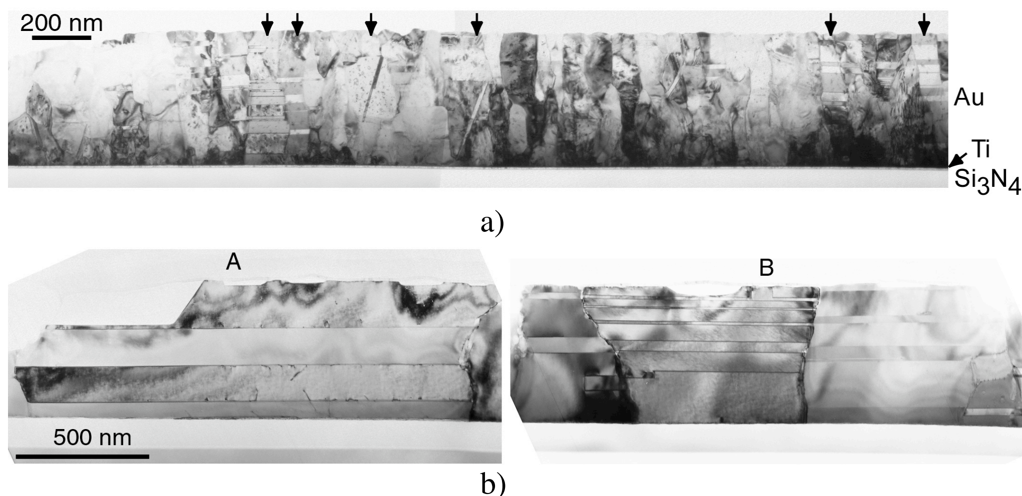


Figure 3: Microstructure of a 500 nm thick gold films : a) as-deposited, b) thermally cycled to 500°C (post-mortem). Note the amount of twinned grains present in the as-deposited film (black arrows) and in grains A and B in the cycled film.

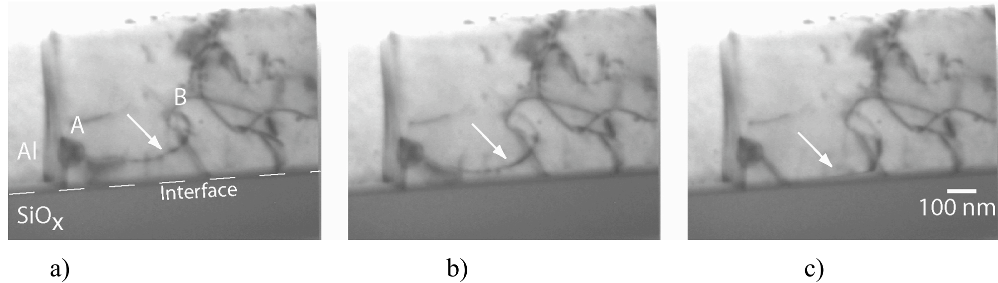
The as-deposited films microstructure (Al, Al(Si,Cu), Cu) always consists of tangled dislocations with a density of the order of 10^{+9} cm^{-2}. The Au films have a columnar grain structure while the Al films, which were deposited at higher homologous temperature, have a "pancake-like" grain structure, where the grains are usually larger than the film thickness. Because of their fine initial microstructure, Au films exhibit a very large grain growth along the interface (Fig. 3).
As seen in Fig. 3b, this grain growth leaves the twin boundaries that are parallel to the interface mostly unaffected. These twins are very stable and form a strong barrier for dislocation transmission. Since they are present in most of the grains, one can assume that, from a channeling dislocation point of view, they reduce the effective thickness of the film. During in-situ experiments, grain growth has not been observed in the case of gold films, probably because of shortened cycles. In the case of Al films, grains were initially large, which explains less extended growth during thermal cycles.

In all cases, dislocation density exhibits a significant decrease throughout thermal cycle. This is particularly visible in the cases of Al and Al alloy films on Si substrates and Cu on sapphire. In cross-section in-situ samples, loss of dislocation occurs rapidly because of the added free surfaces. In Al films, very scarce and ineffective dislocation sources were observed while grain growth was not complete. These sources, generally located at the intersection between the film/substrate interface and a grain boundary appeared however unable to balance the dislocation loss. This resulted in almost dislocation free grains after one or two in-situ thermal cycles. The dislocation activity in Au grains has been observed to be very low up to 600°C and most of the times confined between adjacent twin boundaries. This finding confirms the fact that the parallel twins act as strong barriers for dislocations and impose a channeling path for dislocations that is smaller than the total film thickness. No dislocation sources could be seen in Au films.

On the opposite, Cu films show active dislocations sources that are mainly productive during stress relaxation at room temperature (at the end of a cycle). Those observed so far are single-armed sources, located in the central region of the film, probably anchored on a sessile dislocation segment. Once the unnecessary dislocations are removed (typically after 3-4 in-situ cycles), these dislocation sources are able to compensate the loss of dislocations at the free surfaces. Therefore, in the case of Cu films on sapphire, the dislocation density decreases during the first cycles, reaches a minimum and remains constant during successive cycles thereafter. As explained in the next paragraph, the interface also plays a role in maintaining a low, but constant, dislocation density.

3.2 Dislocation/interface interaction

In the case of crystal/amorphous interfaces, such as Al/oxidized Si, the dislocation/interface interaction is attractive. Fig. 4 shows a dislocation loop expanding under stress towards the SiOx layer in a 550 nm AlSiCu film.
Not only the dislocation is not repelled by the stiffer interface as expected from continuum mechanics calculations [17], but it is also attracted by it, as shown by the acceleration of its motion from figure 4a to 4c. Once at the interface, the dislocation vanishes, probably through atomic rearrangement of the Al/SiOx atomic bonds, and cannot be re-emitted from the interface. This means that the dislocation loss observed in Al, alloyed Al and Au films on Si substrates covered with oxide or nitride not only occurs at the free surfaces, but also at the metal/oxide or metal/nitride interface.



a) b) c)

Figure 4: Al(Si, Cu) 550 nm film during in-situ cross section TEM. Dislocation (white arrow) moving towards the Al/SiOx interface at a) 0s, b) 5.5s and c) 7s. T=170-180°C, heating up.

This phenomenon does not take place in Cu films deposited on sapphire. In this later case, dislocations can also be incorporated into the interface, providing an additional stress exists: rearrangement of incoming dislocations with the existing interfacial network is only observed when pile-ups are formed in Cu, generally at a Franck-Read source. These sources have been found to operate at room temperature, after a thermal cycle was performed. At the tip of the pile-ups, the stress concentration is sufficient to push the first dislocation into the pre-existing network. When the film is heated again, dislocations are re-emitted from the interfacial dislocation network, which plays the role of a dislocation tank, and glide towards the upper free surface of the film. Some of these dislocations can trigger a dislocation source, as described earlier. Contribution of interfacial and source dislocations explains why dislocation density in Cu films on sapphire stabilizes after having reached a minimum. This constant amount of mobile dislocations is able to provide enough plastic deformation to compensate the deformation demanded during additional thermal cycles.

4. DISCUSSION AND CONCLUSION

In-situ TEM on various metallic films has shown that the type of film/substrate interface played a key role in the dislocation microstructure evolution during thermal cycles. Wafer curvature measurements indicate that, in this thickness range, Al and Cu films reach lower yield stresses when deposited on sapphire than when attached to an amorphous diffusion barrier [18]. From a dislocation point of view, this behavior can only result from two mechanisms: either the dislocations see a smaller channeling path in the case of the amorphous coated substrate or their motion is hindered by some intrinsic or extrinsic factor. As stated earlier, forest mechanisms (entangling dislocations) have rarely been observed [11] and never in the experiments reported in the present study. Moreover, the dislocation density remained higher in Cu/sapphire films (e.g. the softer system). In large grains structure, grain boundaries do not provide a significant alternative obstacle. Dynamic aging and impurities could also block dislocation motion, but no evidence of it has ever been found. On the contrary, the motion of dislocations in pure Al films is very similar to what is observed in pure bulk Al and in-situ TEM is a very sensitive technique to unveil this type of interaction. For a given film thickness, a smaller channeling path could result from elastic interaction with interfacial dislocations or from images forces. These images forces increase with the shear modulus ratio between the substrate and the film, e.g. a stiffer substrate will provide higher repulsion for dislocations in the film. Also, interfacial dislocations are only present in the case of sapphire substrate. Thus, the channeling path is shorter in the case of a sapphire substrate and should therefore lead to a stronger film. Experiments prove that the contrary happens.

What the recent wafer curvature literature and the present in-situ TEM observations suggest is that dislocations-based mechanisms may have a very modest impact on the thin film mechanical behavior when a crystal/amorphous interface is considered. Threading dislocations have an attractive interaction with the amorphous interface, resulting both in a limited confinement and in a dislocation density loss. The remaining dislocations cannot account for the plastic strain observed in subsequent cycles. On the opposite, these mechanisms can account well for the yield stress increase in the case of metallic films with a crystal/crystal interface. This interface provides both the expected confinement for threading dislocations and dislocation renewal to balance the loss at the free surface. Fresh dislocations are nucleated directly from the interfacial network or through dislocations sources located in the film thickness. The link between these last two mechanisms is under investigation.

5. ACKNOWLEDGMENTS

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6. REFERENCES

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