PLASTICITY AND FRACTURE OF METALLIC THIN FILMS

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ABSTRACT

Micrometer- und sub-micrometer thick metallic films on substrates reveal mechanical properties, which deviate from the corresponding bulk metals. For example, Cu and Al films can reach exceptionally high flow stresses of several hundred MPa, while brittle NiAl films reveal fracture stresses of upto 2000MPa. A basic understanding of the size effects controlling the mechanical properties of thin films will provide new insight into materials behavior in small dimensions and may be relevant in improving the reliability of miniaturized devices suffering from high internal stresses. In order to identify the deformation mechanisms and correlate them with the microstructure and film thickness, thermally strained Al, Cu and NiAl films were analysed. Polycrystalline films were deposited by magnetron sputtering on nitrided and/or oxidised (001) Si substrates, while epitaxial Al and Cu films were grown on (0001) α-Al_2O_3 single crystals. Flow stress measurements of the epitaxial films, which were obtained from substrate-curvature tests, follow predictions from a dislocation-based model. In contrast, the flow stresses measured for polycrystalline Al and Cu films are much higher than predicted values. A similar behavior is observed for intermetallic NiAl films. However, Al-rich NiAl films exhibited crack formation during thermal straining. The fracture stress increased with decreasing film thickness, but the fracture toughness of the Al-rich films remained constant independent of the NiAl film thickness. The flow stress and cracking behavior of the films will be discussed in terms of existing theories for plasticity and fracture of thin metallic films, and under consideration of recent in situ transmission electron microscopy studies.

1 INTRODUCTION

Mechanical stresses in small-scale structures, such as thin metallic films on substrates or oxidation and corrosion resistant coatings often reach values of several hundred MPa after processing or during service [e.g.1-7]. These high stresses can in turn induce defects that cause failure of complete devices and coating structures. The mechanical stresses usually increase with decreasing geometrical and/or microstructural dimensions, making stress-induced materials failure of film/substrate systems increasingly likely especially with the present trend of miniaturization.

In the present overview, recent advances in thin film plasticity and channel cracking are summarized using examples of Al [3,7-9], Cu [4,10] and NiAl [6] thin film studies. Al and Cu represent thin film materials frequently used in microelectronic devices and microelectro-mechanical systems, while NiAl-based alloys are typical coating materials for the protection of turbine blades against high temperature oxidation. Polycrystalline NiAl, Al, and Cu films were deposited by magnetron sputtering on nitrided and/or oxidised (001) Si substrates, while epitaxial Al and Cu films were grown on (0001) α-Al_2O_3 single crystals. The film thicknesses ranged between 100nm and 3000nm. The mechanical behavior of the films was deduced from thermal straining experiments using a substrate-curvature technique [11]. Focused ion beam (FIB) microsccopy and in-situ transmission electron microscopy (TEM) studies were performed on plan-view and cross-sectional specimens to obtain insight into fracture processes, dislocation mechanisms, and diffusion related phenomena.

2 EXPERIMENTAL

Polycrystalline Al [7], Cu [4], and NiAl [6] films were grown on (001)-oriented Si substrates with a 50nm thick amorphous SiN_x and/or 50nm thick amorphous SiO_x diffusion barrier. The chemical composition of the NiAl films was Al rich with Al contents of 52.2 and 52.4at% Al. Furthermore, epitaxial Al [8] and Cu [3] films were deposited on (0001)-oriented α-Al_2O_3 substrates. This allowed the effects of film thickness and grain size on thin film plasticity to be investigated separately. All films were deposited at ambient temperature. Al and Cu films were usually annealed directly after film deposition under ultrahigh vacuum conditions in the growth chamber for at least 10min, while the NiAl films were frequently annealed ex situ in a protective N_2 atmosphere and the stress evolution during thermal cycling was measured. Thermal cycling resulted in maximum film strains of 0.5 to 0.9%. The typical microstructure of the various films are summarized in Fig. 1.

3 RESULTS AND INTERPRETATION

3.1 Plasticity in thin Al and Cu Films

FIB studies of the epitaxial Al and Cu films revealed two twin-related growth domains. Epitaxial Al films formed bicrystals (Fig. 1a) exhibiting a $\{111\}_{Al} \pm \langle 110\rangle_{Al} \parallel (0001)_{Al2O3} \langle 10\bar{1}0\rangle_{Al2O3}$ orientation relationship [8]. Epitaxial Cu films grew mainly with a $\{111\}_{Cu} \pm \langle 112\rangle_{Cu} \parallel (0001)_{Al2O3} \langle 10\bar{1}0\rangle_{Al2O3}$ orientation relationship [3]. In this case, deviations from the in-plane orientation relationship of upto 10 degrees were observed. FIB studies of the polycrystalline Al (Fig. 1b) and Cu films revealed columnar grains that extend over the complete film thickness. Selected area diffraction studies in the TEM revealed a {111} fibre texture, which was confirmed by X-ray diffraction investigations.

The thermal stress evolution of the Al and Cu films was found to depend strongly on the microstructure, as illustrated by Figs. 2. While epitaxial films exhibit nearly constant flow stresses in compression and in tension (Fig. 2a), polycrystalline films show a significant increase in the tensile flow stress upon cooling (Fig. 2b). In contrast, the compressive flow stress values of polycrystalline and epitaxial films are similar.

Cross-sectional in-situ TEM studies of epitaxial Al and Cu films indicated that dislocations can channel through the film, laying down an interfacial segment at the metal/Al_2O_3 interface [3,7,9]. The same constraining mechanism is also present at $\Sigma 3$ {111} twin boundaries oriented parallel to the film surface as observed in a 600nm thick epitaxial Cu film (Fig. 3a). A similar dislocation mechanism was proposed by Nix and Freund [e.g. 1] and calculated flow stresses from their simple dislocation-based model amount to 75MPa and 105MPa for 600nm thick Al and Cu films, respectively. While this dislocation channelling mechanism [1] can explain and predict the thickness dependence of the flow stress in epitaxial Al and Cu films [3], it significantly

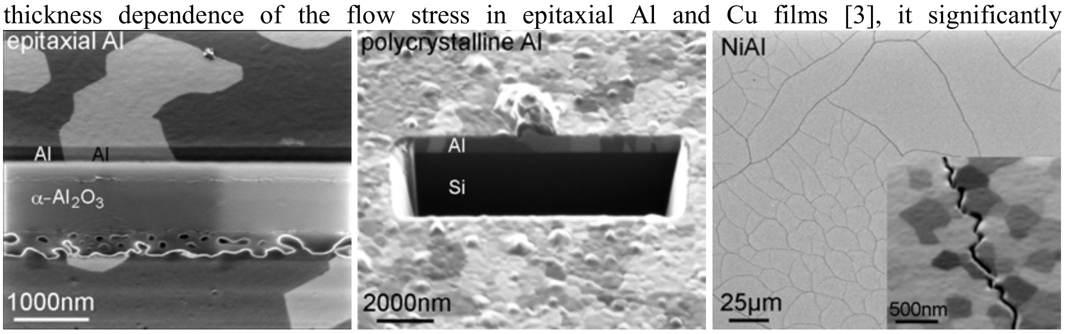

Fig. 1: FIB images of (a) a 350nm thick epitaxial (bicrystal) Al film grown on a (0001)-oriented α-Al_2O_3 single crystal [8], (b) a 1000nm thick polycrystalline Al film grown on an oxidised (001) Si substrate [3], and (c) a 1500nm thick Al rich NiAl film (52.2at% Al) on a nitrided (001) Si substrate. Note that cracks are present in the Al rich NiAl film, which run mainly along NiAl grain boundaries (see enlarged inset in (c)) [6].

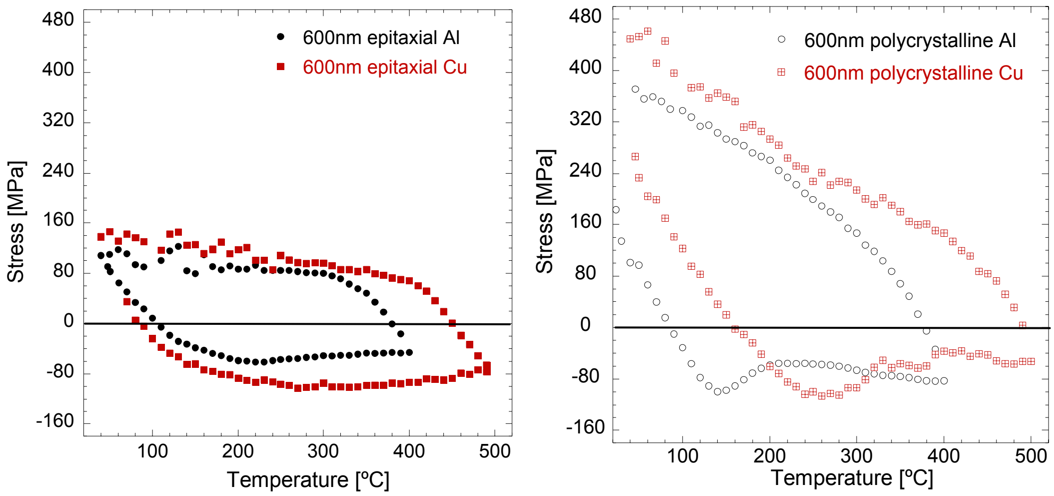

Fig. 2: Thermal stress evolution of 600nm thick (a) epitaxial and (b) polycrystalline Al and Cu films, respectively, as measured with a substrate-curvature technique [3].

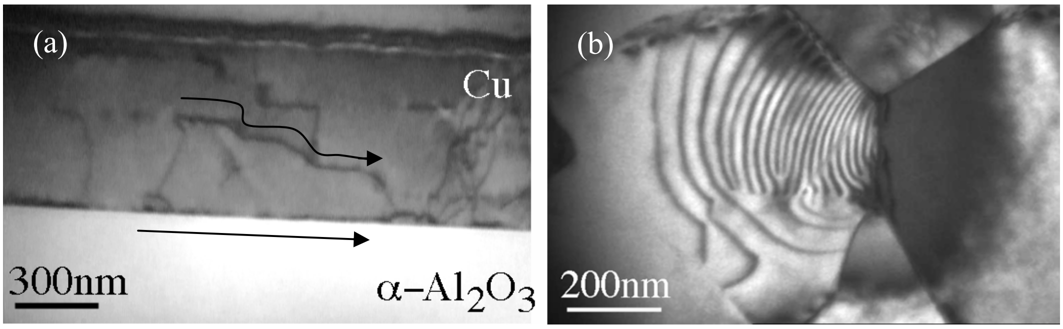

Fig. 3: (a) A threading dislocation gliding on an inclined $\{111\}_{Cu}$ plane gets constrained between a Cu twin boundary and the film/substrate interface. The advancing dislocation is forced to deposit interfacial dislocation segments at both interfaces (see arrows) [3]. (b) Parallel glide dislocations [4] emitted from a grain boundary in a 150nm thick Cu film during cooling from 500°C to 40°C.

underestimates tensile flow stresses in polycrystalline Al and Cu films.

However, the moderate compressive stresses in polycrystalline films can be related to diffusional creep mechanisms. In polycrystalline Al films hillock formation (see Fig. 1b) by transport of matter along the grain boundary relaxes the compressive film stresses upon heating [e.g.14]. Likewise, stress relaxation by grain boundary diffusion occurs in polycrystalline Cu films at elevated temperatures [12,13]. In contrast to Al, Cu does not form a passivating surface oxide. Thus grain boundary diffusion is coupled with surface diffusion, preventing the formation of hillocks in Cu thin films. However, for polycrystalline metal films on substrates the film/substrate interface retards diffusion compared to Coble creep and the resulting grain boundary diffusion mechanism is termed "constrained diffusional creep" [12]. Constrained diffusional creep can cause localized shear stresses on the (111) plane parallel to the substrate with substantial dislocation activity, as found by a series of plan-view in-situ TEM studies of polycrystalline Cu films on

SiN_x/Si substrates [4,10] (Fig. 3b). These dislocations are thus referred to as “parallel glide” dislocations [4], and were observed for Cu film thicknesses below 400 nm and down to 50 nm.

3.1 Channel Cracking in thin Al rich NiAl Films

The Al rich NiAl films consisted solely of equiaxed grains of ordered β-NiAl with a mixed {111} and {211} texture. Upon cooling from temperatures exceeding 600°C the films developed a network of microcracks (see Fig. 1c). The cracks propagated predominantly in an intergranular manner. On a larger scale, the cracks are preferentially along <110> directions of the Si substrate. Cross-sectional FIB studies revealed that cracks extended over the entire film thickness and penetrated into the Si substrate [6].

Thermal stress measurements during cooling revealed the onset of cracking by the occurrence of a stress drop (see Fig. 4a). The fracture stress of the films increased with decreasing film thickness (Fig. 4b). A good agreement is found with existing models of thin film fracture [15-17] in which the film thickness corresponds to a critical defect size. The fracture toughness of the Al-rich NiAl films was determined to be in the range of 2.2 to $2.8 MPam^{1/2}$ [6]. Contrary to the fracture stress, the fracture toughness showed no distinct dependence on the film thickness [6].

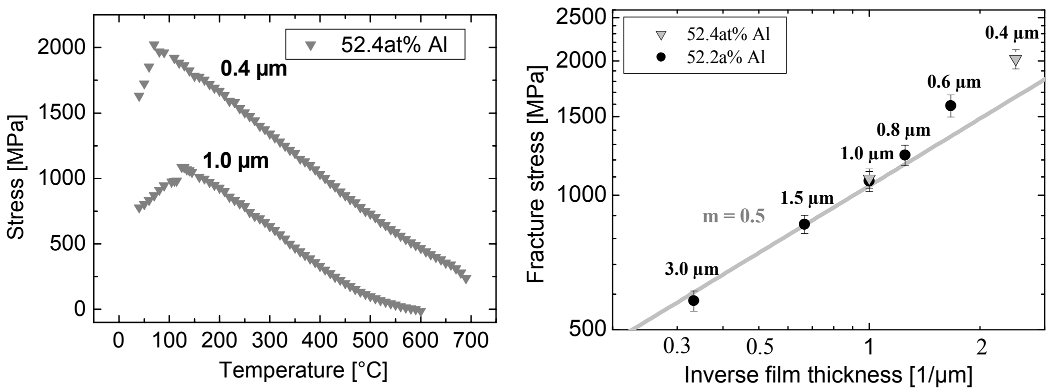

Fig. 4: (a) Stress evolution on cooling from 600 or 700ºC to 40ºC for Al rich NiAl films. A stress drop occurs indicating fracture [6]. (b) The measured fracture stress values of NiAl films with Al contents of 52.2 and 52.4at% are shown as a function of the inverse film thickness (double logarithmic plot). Note that the fracture stresses follow a predicted slope of 0.5 indicating that the film thickness corresponds to the critical defect size [6].

4 SUMMARY

The combination of wafer-curvature measurements with in-situ TEM experiments, as well as the comparative study of epitaxial and polycrystalline Al and Cu thin films, have shed new light on the basic plasticity mechanisms in thin metallic films: Heteroepitaxial Al and Cu films on (0001) α-Al_2O_3 substrates exhibit nearly ideal elastic-plastic stress-temperature evolution during thermal straining. Dislocations advancing through the film leave behind interfacial dislocation segments at the film/substrate interface. Dislocation motion gets increasingly difficult by the presence of interfaces acting as obstacles. In polycrystalline Al and Cu films, grain boundary diffusion relieves stresses at elevated temperatures. Hillocks form in Al films under compressive stresses, while constrained diffusional creep relaxes stresses at grain boundaries in unpassivated polycrystalline

Cu films at elevated temperatures. If no interface diffusion or sliding occurs, constrained diffusional creep produces stress inhomogeneities and induces parallel glide. Parallel glide dislocations are emitted from grain boundaries and glide on a (111) plane close to the Cu/a-SiN_x interface. However, flow stresses at room temperature always exceeded those of their epitaxial counterparts. Although diffusional stress relaxation processes are active in polycrystalline films, the additional constraint by grain boundaries on the motion of threading dislocations surpasses the stress relaxation by grain boundary diffusion, most notably during cooling from elevated temperature to room temperature.

In contrast to the ductile Al and Cu films, channel cracking occurred in Al rich NiAl films. The fracture stress of these NiAl films was found to increase with decreasing film thickness. However, the fracture toughness remained constant at 2.2 to 2.8MPam$^{1/2}$ independent of film thickness. This result demonstrates that the film thickness corresponds to a critical defect size as predicted in existing models of thin film fracture.

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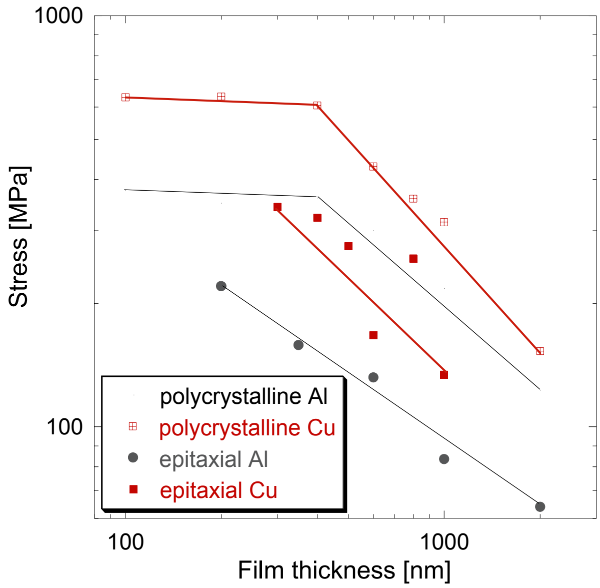

tress measurements of the films were obtained from substrate-curvature tests., follow predictions from a dislocation-based model. In contrast, the flow stresses measured for polycrystalline Al and Cu films are much higher than predicted values and reveal a plateau in room temperature flow stress for ultrathin films. A similar behavior is observed for stoichiometric and Ni-rich intermetallic NiAl films. However, Al-rich NiAl films exhibited crack formation during thermal straining. The fracture stress increased with decreasing film thickness, but the fracture toughness of the Al-rich films remained constant independent of the NiAl film thickness.

The flow stress and cracking behavior of the films will be discussed in terms of existing theories for plasticity and fracture of thin metallic films, and under consideration of recent in situ electron microscopy studies.