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In-situ TEM straining experiments of Al films on polyimide using a novel FIB design for specimen preparation

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Abstract In-situ transmission electron microscopy (TEM) straining experiments are tedious to perform but give invaluable insight into the deformation processes of materials. With the current interest in mechanical size-effects of nanocrystalline materials and thin metallic films, in-situ tensile testing in the TEM is the key method for identifying underlying deformation mechanisms. In-situ TEM experiments can be significantly simplified using well-designed specimens. The advantages of a novel focussed ion beam design and first in-situ straining results of 500-nm thick single-crystalline Al films on polyimide are reported and compared to conventionally prepared Al films on polyimide.

Introduction

Dislocation-based plasticity is still of vital research interest in materials science in order to understand deformation processes, creep properties, fracture behaviour, and fatigue phenomena. For bulk materials, however, debates frequently arose whether the dislocation mechanisms identified by transmission electron microscopy (TEM) using a few hundred nanometer thick specimens are the same as in the corresponding bulk material. This concern is overcome for TEM experiments on materials with small (submicrometer) geometrical and/or microstructural dimensions such as thin films and nanocrystalline materials. As a consequence, in-situ TEM techniques are the methods of choice for identifying deformation mechanisms in small-scale structures. Especially, mechanical size-effects of materials with the general trend of ''smaller is stronger'' [1–3] demand in-situ deformation experiments to clarify the underlying mechanisms. This has led to the development of novel and quantitative in-situ TEM techniques such as indentation tests [e.g., 4, 5] and micro-electro-mechanical system (MEMS) based straining devices [6], which allow recording of the load–displacement curves of the sample during TEM observation. Such quantitative in-situ TEM experiments are not straightforward to perform and currently limited to a few laboratories in the world.

In contrast, classical in-situ TEM deformation experiments by heating and/or straining, which do not provide a stress–strain curve of the analysed material but yield valuable information on e.g., local stresses and dislocation velocities, are more widespread and standard since the 1970s. They are usually aimed at accompanying quantitative ex situ experiments in order to reveal the active dislocation mechanisms. However, the sample preparation is tedious, especially for mechanical straining tests in the TEM, and these experiments often fail because the region of interest remains unstrained, bends or cracks.

For conventional TEM studies the focused ion beam (FIB) microscopes have brought many benefits in sample preparation. The main advantages are short preparation

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times of typically less than day, a homogeneous TEM sample thickness, and the possibility of a site-specific preparation. While numerous articles describe TEM sample preparation routes by FIB [e.g., 7–9] very little use of the FIB for making in-situ straining TEM samples has been made so far [10].

In this study the FIB is used to design straining samples of 500-nm thick single-crystal Al films on polyimide. The performance of the FIB-shaped samples are compared to conventionally prepared straining samples of 500-nm thick single-crystal and polycrystalline Al films on polyimide using dimpling and Ar-ion milling. The dislocation mechanisms in the Al films are discussed and compared to results in the literature.

Experimental details

Deposition of Al films

Epitaxial and polycrystalline Al films were grown by magnetron sputtering on (100) oriented single-crystal NaCl substrates. For the growth of epitaxial films commercial epi-polished substrates were used (Crystec, Berlin), while polycrystalline Al films were grown on less well-defined NaCl substrate surfaces obtained by cleavage from a thick crystal rod. Sputter-deposition of the 500-nm thick films was performed at nominally 300 °C in a commercial deposition system (DCA Instruments, Finland) with a base pressure of better than 10^{-8} Pa. The elevated temperature during deposition led to columnar grains for the polycrystalline films and helped to anneal out point defects introduced during sputter deposition, since point defects alter the mechanical response of thin metallic films [11]. Subsequently, the epitaxial and polycrystalline Al films were spin-coated with a ~9-µm thick polyimide layer using precursors from DuPont (USA). The polymer/film/NaCl samples were baked at 330 °C for several hours in a nitrogen atmosphere to cure the polymer coating.

Fabrication of the tensile testing specimens

The samples for in-situ straining experiments in the TEM consisted of two components: a Cu support which fits into the straining stage, and the actual TEM sample which was glued to the Cu support [12] either before or after the NaCl substrate was removed by de-ionised H_2O. The rectangular Cu supports were shaped by spark erosion; they are 3 mm wide, 6 mm long, about 0.1 mm thick and have 1-mm diameter holes at both ends (see Fig. 1a) in order to fit exactly into the straining stage. The centre of the support contains a 1×2 mm^2-sized rectangular hole on to which the actual TEM sample—in this study a polyimide/Al stripe of ~3 mm length and less than ~2 mm width—was glued with the region of interest positioned above the hole in the support. A two-component glue (e.g., from Gatan) or a conventional superglue was used to fix the TEM sample on the Cu support. Two kinds of TEM sample processing routes were compared: a FIB design and a conventional route based on dimpling and Ar-ion milling.

The FIB design started with cutting a ‘‘dog-bone’’ shape into the straining sample by Ga-ion milling using a FIB microscope (FEI 200 xP). At both sides of the 3×2-mm^2 polyimide/Al stripe trapezoids of several hundred micrometer length were removed (Fig. 1b) using a Ga-beam current of 11.5 nA at an accelerating voltage of 30 kV. In our case with a polyimide thickness of 9 µm the cuts on each side of the sample took about 5 h. In the centre region a square of up to 100×100 µm^2 in dimension was milled down to a remaining thickness of less than ~3 µm using a beam current of 11.5 nA at an accelerating voltage of 30 kV (Fig. 1b) in order to obtain an electron transparent area of constant thickness *without perforation* (Fig. 1c). This step took about 5 h. The Ga-ion milling time was calibrated for the applied parameters by cutting test structures in a polyimide reference sample. Finally, a polishing step with a beam current of 2.7 nA was applied to the 100×100 µm^2 centre region. The maximum total FIB time for designing the straining sample presented in Fig. 1 was about 17 h. Most of the Ga-milling was performed automatically overnight. The milling time can be minimized to several hours by using smaller dimensions for the electron transparent region and for the trapezoids. This did not cause any drawbacks for the in-situ straining experiments as indicated by TEM straining tests of samples with a 50×50 µm^2 centre region and trapezoids of 100 µm in length.

While the FIB technique was used in this study solely for the epitaxial Al films grown on (100) NaCl crystals, a conventional TEM sample preparation route was used for all the polycrystalline Al films and a few single-crystalline films. The polyimide-coated polycrystalline Al films were also cut into stripes of ~3 mm in length and less than 2 mm in width using a razor blade and dissolved from the NaCl substrate with de-ionised H_2O. The stripes were gently dimpled from the polyimide side (only 1–2 rounds) and then glued to the Cu support. Finally, the tensile testing samples were Ar-ion milled from the polyimide side at 6 kV in a Gatan Duo mill with liquid-N_2 cooling until a small hole appeared in the dimpled area. All tensile test samples were carbon coated from the polyimide side to prevent charging in the TEM.

TEM straining experiments were performed at a 200 kV TEM (Jeol 2010) using a Gatan single-tilt straining holder. The in-situ experiments were recorded on DV-Cam using a TV-rate camera. Additionally, negatives were recorded at certain strain levels. The

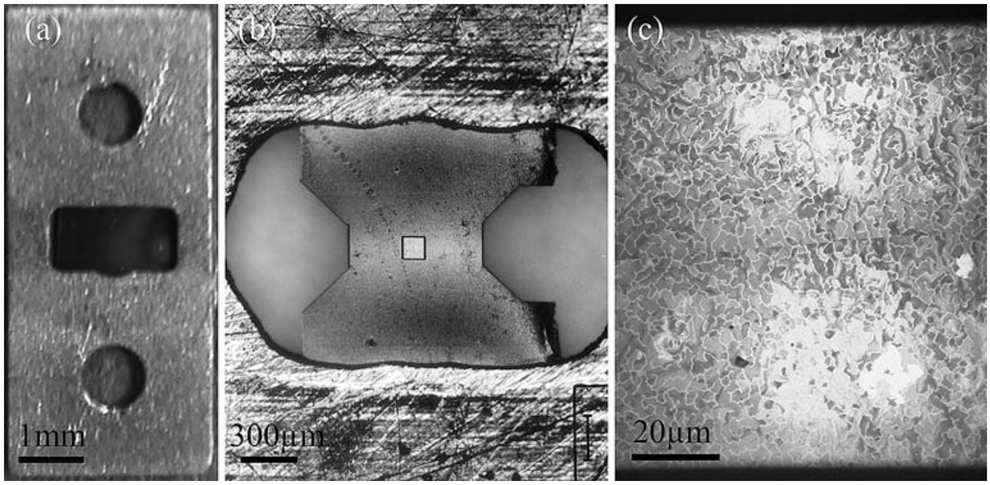

Fig. 1 (**a**) Optical micrograph of a Cu support. The central square-shaped hole (''window'') is cut by spark erosion. The TEM specimen is glued on the Cu support and stretches over the ''window''. (**b**) Trapezoids are cut out at the sides of the specimen and a 100 × 100-µm^2 large region is milled to electron transparency using the FIB microscope. (**c**) TEM low-magnification image of the electron transparent region. The contrast features in the image stem from thickness inhomogeneities of the Al film which grows three dimensionally on the salt substrate. The same contrast was also observed for dimpled and Ar-ion milled samples

experimental set-up does not permit stress–strain curves of the Al films to be obtained, but the FIB design provided a simple method to localize the region of interest during a TEM straining experiment. This in turn helped to detect the deformation processes during an in-situ deformation test which are often obscured in a conventionally prepared TEM straining sample by motion of bend contours and difficulties in localizing the strained area due to perforations (holes) in the sample.

TEM observations and in-situ straining results

All tensile testing samples contained dislocations at the beginning of the TEM experiments. Most likely, dislocations have already formed during growth of the Al films on the NaCl substrate and during the annealing treatments as a consequence of the differences in thermal expansion coefficients of polyimide, Al, and NaCl.

The epitaxial Al straining sample is aligned close to a $\langle 100 \rangle$ Al zone axis without any tilting in the TEM (Fig. 2a) indicating that the Al film grew with the (100) plane on top of the (100) oriented NaCl substrate [13]. The epitaxial Al films were found to be single crystal within the electron transparent regions made by the FIB.

Initially, dislocation motion was observed by local beam heating, which stopped after irradiating the same region for some tens of seconds. Subsequently, the sample was mechanically strained along a $\langle 100 \rangle$ Al tensile axis. Upon straining the onset of plasticity can be seen by significant dislocation activity in the electron transparent centre region. Dislocations channel mainly along $\langle 110 \rangle$ Al directions and lay down an interfacial dislocation segment at the Al/polyimide interface (Fig. 2b). Upon straining dislocation bursts frequently occurred with the individual dislocations travelling too fast to analyse dislocation motion and dislocation interactions by a frame-by-frame inspection of the videotape. With a typical field of view of

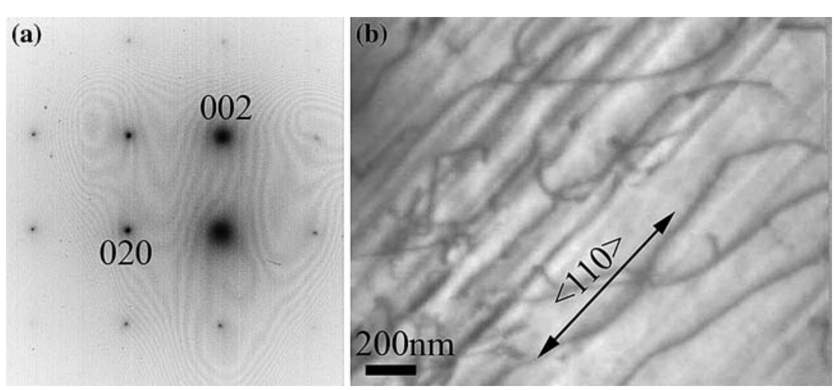

Fig. 2 (**a**) TEM diffraction pattern of Al in $\langle 100 \rangle$ zone axis. (**b**) Bright-field TEM image recorded with g_{002}. Note the interfacial dislocation segments along $\langle 110 \rangle$ Al directions

2.5 μm, dislocations travelling faster than ~62.5 μm/s are not detected within a single video frame. Between the straining periods relaxation by jerky dislocation motion dominates. This behaviour is often accompanied by local dislocation re-arrangements. Dislocation nucleation events are difficult to spot, but in two cases clear evidence for a Frank-Read source type of dislocation emission was found with one of the sources emitting more than four dislocations. Fig. 3a and b show the source briefly before emitting a dislocation and directly after emission of the dislocation. The minimum size of this source measured from the distance between the anchoring points visible in Fig. 3b is estimated to be ~200 nm ± 20 nm considering an error of ±4° in tilt accuracy and correcting for the projected distance by assuming that the dislocation source is lying on an inclined {111} Al plane. The distance of 200 nm for the dislocation source size corresponds to a shear stress τ of

$$\tau = Gb/L \approx \frac{26\,\mathrm{GPa}\ 0.286\,\mathrm{nm}}{200\mathrm{nm}} = 37\,\mathrm{MPa} \qquad (1)$$

for the operation of the source [14]. G = 26 GPa is the shear modulus of Al [14], b = 0.286 nm the Burgers vector, and L = 200 nm the experimentally deduced distance of the anchor points of the source.

The Ar-ion milled samples reveal initially thermally driven dislocation motion under the electron beam similar to the FIB designed epitaxial Al films. Dislocations channel along $\langle 110 \rangle$ Al directions and form stable interfacial dislocation segments at the film/polyimide interface. Upon mechanical straining the onset of plastic deformation was difficult to detect. The Ar-ion milled sample always possessed several holes (see Fig. 4) and were inhomogeneous in thickness. This led to stress inhomogeneities and only some localized regions of the electron transparent area were strained. These regions were hard to find and had to be searched for by looking at different places of the sample during straining. Usually, a crack opened up in the polyimide layer, which was bridged by the intact Al film (Fig. 5). In this region strong dislocation activity in the Al was detected.

The epitaxial and polycrystalline Al films on polyimide finally failed by cracking with the cracks running along $\langle 100 \rangle$ Al directions in a zig-zag manner (Fig. 6) independent of the preparation route. Ahead of the crack high dislocation activity was noticed with local thinning of the Al in front and adjacent to the advancing crack.

Discussion

A simple and novel concept to design TEM tensile testing samples using the FIB microscope has been successfully implemented. The advantage of this method lies in the fact that the straining samples have a homogeneous thickness in the electron transparent region. The sample design makes it easy to localize the onset of dislocation-based plasticity during straining experiments. Furthermore, if further thinning is required to improve the electron transparency of the sample this can be achieved with a subsequent conventional Ar-ion milling as tested during the experiments. In contrast to the method described by Field and Papin [10] no lift-out technique and accurate positioning of a tiny, micron-sized specimen is required. The FIB-based fabrication process is an alternative if a lithographic process as implemented e.g., by Hattar et al. [15] is not available. In principle the FIB design can be also used to make TEM straining samples from bulk materials. However, care has to be taken that implanted Ga-ions will not affect the mechanical response. In the present study no point defect clusters were observed in the Al films indicating that the remaining polyimide thickness was sufficient to prevent noticeable Ga implantation into the Al.

The straining experiments revealed that dislocations channelling along $\langle 110 \rangle$ directions lay down interfacial

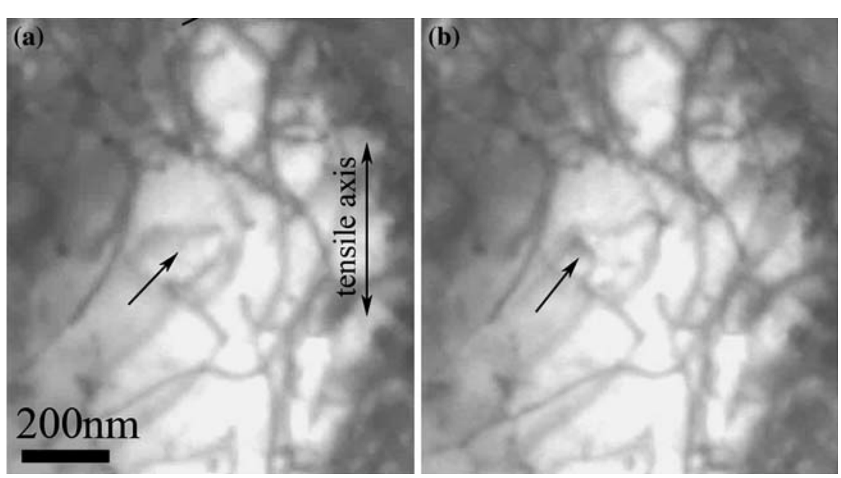

Fig. 3 Dislocation emission from a source operating during straining. (**a**) The video frame shows the source at maximum diameter (*arrow*) and (**b**) directly after dislocation emission (*arrow*)

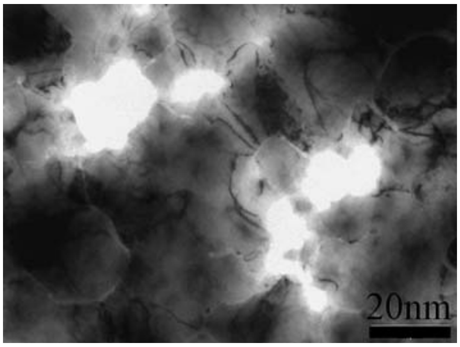

Fig. 4 Small holes adjacent to a larger hole (not visible in this figure) in a polycrystalline Al film on polyimide after Ar-ion milling

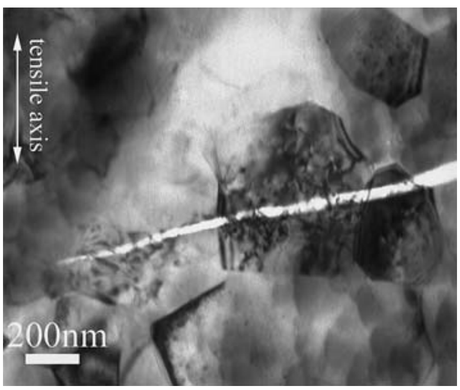

Fig. 5 TEM image of a conventionally prepared straining sample. A crack has formed in the polyimide substrate. The Al film on top is still intact as can be seen by features in the Al grains ''stretching'' over the crack

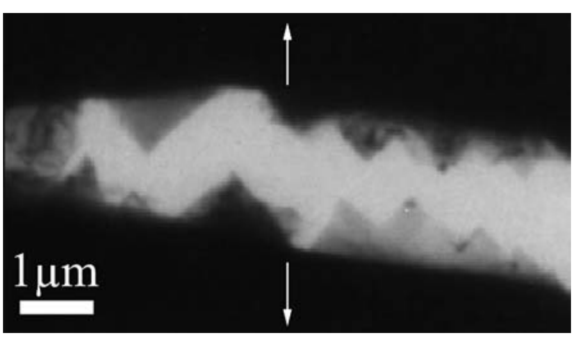

Fig. 6 Video frame of a crack, which has formed in a FIB shaped straining sample during deformation. The arrows indicate the ⟨100⟩ straining direction. The crack runs zig-zag along ⟨110⟩ Al directions

dislocations. This mechanism has been proposed by Freund [16] and Nix [1] as an explanation for the experimentally observed increase in flow stresses for decreasing film thickness of metallic films on substrates. While this mechanism has been known to operate in semiconductor films on substrates, it was only recently observed for single-crystal metal films on ceramic substrates [17, 18]. In contrast, metal films on amorphous interlayers revealed in several cases that interfacial dislocation segments are not stable or do not exist at an interface between a crystal and an amorphous template [19–21] since it can behave like a free surface if the interfacial bonding allows a spreading/relaxation of the dislocation core. It seems that the bonding between Al and polyimide is strong enough to prevent a noticeable disappearance of interfacial dislocations at room temperature.

The occurrence of dislocation bursts in the single-crystal Al films has been also observed in in-situ thermal straining experiments of (100)-oriented epitaxial Al films on Si [22] and in (111)-oriented Al films on α-Al_2O_3 substrates [23]. The observation of a dislocation source operating several times with dimensions between 1/3 and 1/2 of the film thickness is in agreement with a dislocation-source model for thin-film plasticity developed by von Blanckenhagen et al. [24]. However, further studies with different film thickness are necessary to verify this model or to propose alternative mechanisms causing a size-dependent flow stress for thin films.

The only comparison that can be drawn from the present in-situ experiments of single-crystal and polycrystalline Al films concerns the fracture behaviour. Both types of films fail by ductile fracture with cracks running along ⟨110⟩ directions in accordance with the dislocation glide directions. In the fractured region the Al is thinned down by substantial dislocation plasticity. This mechanism is in agreement with failure of thin films already reported in the 1970s [see e.g., 13], but different from nanocrystalline Al with a film thickness and grain size of ≤130 nm, where semi-brittle fracture with cracks running along grain boundaries was observed [15]. It may be worth noting that first in-situ straining experiments on a 60-nm thick single-crystal Au film on polyimide also revealed ductile fracture [Dehm and Legros unpublished results] similar to the 500-nm thick Al films of this study.

Future straining experiments with different film thicknesses have to be performed to show, whether a ductile-to-brittle transition occurs for ultrathin face-centred cubic metal films.

Summary

A novel FIB design of TEM straining samples was demonstrated for Al films on polyimide to simplify the in-situ experiments. A central ''observation window'' is milled into the polyimide substrate to obtain a well-defined electron-transparent region in the sample. During straining the onset of plasticity in the Al films was easily revealed and dislocations channelling along ⟨110⟩ Al directions were observed. The presence of interfacial dislocations is in agreement with model predictions in the literature for single-crystal films on substrates. First results on a dislocation source indicate a source size of 2/5 of the film thickness. Ductile fracture occurred with cracks running in a zig-zag along $\langle 110 \rangle$ Al directions. It can be concluded from the present study that the tedious search for the region of interest and artefacts introduced by inhomogeneous sample thickness of conventionally dimpled and Ar-ion milled straining samples are easily avoided with the FIB design.

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