# SIZE EFFECT ON FRACTURE IN THIN COPPER/TANTALUM BILAYERS ON POLYIMIDE SUBSTRATES

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ABSTRACT

Both Cu and Ta are established materials for integrated circuits as interconnect and barrier material, respectively. With the advent of polymeric electronics interconnects are being built on rather compliant materials in comparison to silicon wafers. On silicon wafers metal lines were subject to up to 0.5% of total strain due to the mismatch in coefficients of thermal expansion between metal and substrate and the maximum temperatures, they were exposed to during processing. For polymeric substrates design rules have been established that require a maximum total strain of up to 10%. In this study we measured stress/strain curves of Cu/Ta bilayers on polyimide substrates utilizing a recently developed *in situ* synchrotron technique, which will also be introduced in this paper. The Cu thicknesses ranged from 1 µm down to 20 nm, whereas the Ta thickness was always ten nanometers. All experiments were carried out at room temperature. As a reference also single Cu layers were investigated. We found that Cu/Ta samples with a Cu film thickness below 300 nm showed a sudden stress decrease at a total strain of about 2.5%. This stress drop could be attributed to fracture of the entire film system caused by cracks in the Ta layers. Single Cu layers showed no cracking. The stress drop was quantified using analytical models for the stress field around crack patterns in thin films. The size effect will be addressed in terms of fracture mechanics.

## 1 INTRODUCTION

Metals in micro- and nanocomponents are usually subject to very large mechanical stresses. These stresses arise from deposition, microstructural changes, thermal mismatch, or external loading. In many cases stresses are partly relaxed by yielding or creep, both of which are strongly influenced by the dimensions of the components. Most of the research to date has focused on the understanding of deformation mechanisms in thin metal films on stiff substrates [1-6].

Recently, flexible electronic circuits, which have to sustain several percent of total strain, have gained widespread interest for numerous applications including flexible displays [7] and wearable electronics [8]. Therefore completely new processing routes and design solutions have to be developed. The embedding of conductive, metal particles in an elastomer, such as silicone could be a solution [9]. Another possibility is the use of flexible substrates, i.e. polyimide or PDMS and the application of standard metallization systems. For this approach it is important to ensure good adhesion between the metal layers and the polymeric substrate. The maximum total strain of such a structure is limited by the extensibility of the metal film. This can be enhanced by using microfabricated tortuous wires encased in a polymer [10]. Such structures are able to reversibly accommodate linear strains of up to 50% while maintaining conductivity.

Here, we applied a recently developed novel synchrotron X-ray diffraction technique to characterize the evolution of isothermal mechanical stresses during *in situ* tensile tests in ultrathin polycrystalline films on a polymeric substrate [11, 12]. With this method the influence of temperature and time as well as the influence of different cap- and interlayers

on the deformation mechanisms in ultrathin metal films can be investigated. Within the scope of this paper we investigated the deformation behavior and microstructural changes of different Cu/Ta and Ta/Cu/Ta thin film systems during isothermal tensile tests up to total strains higher than 5%.

## 2 EXPERIMENTAL

All samples were prepared by DC magnetron sputtering on polyimide substrates. The substrates were cleaned by ion bombardment in the chamber prior to deposition to ensure good adhesion. The Ta layer thickness was kept constant at 10 nm, whereas the Cu thickness was varied between 20 and 3000 nm. Two magnetron sources were used inside the chamber to avoid a vacuum break between the subsequent deposition of two different layers.

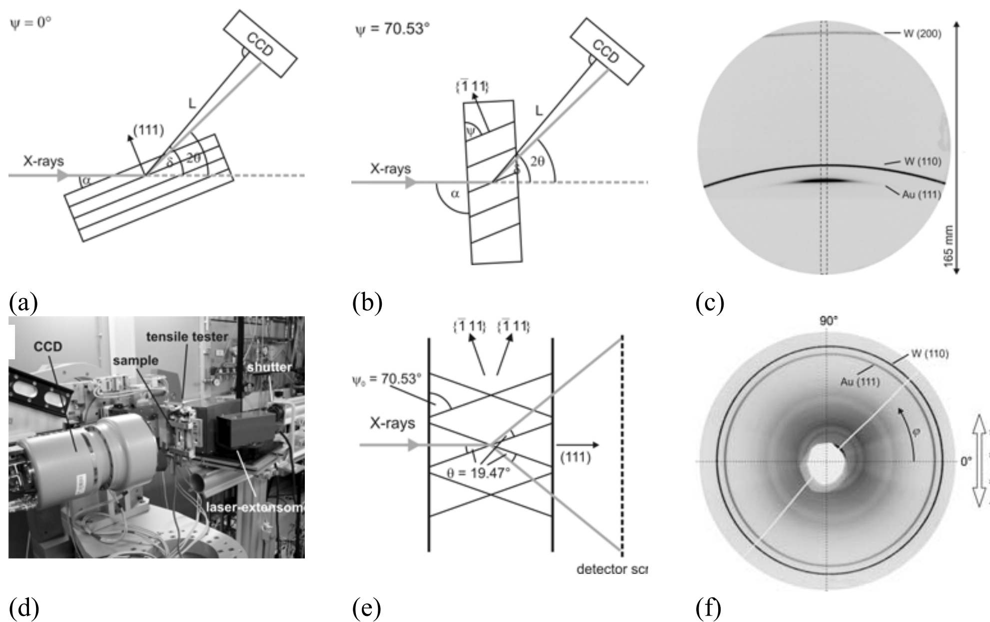


Figure 1 (a)-(b): Diffraction geometries for the "$\sin^2\psi$ method": (a) $\psi = 0°$, (b) $\psi = 70.53°$; (c): Diffraction pattern showing partial Debye-Scherrer rings of a 40 nm Au film and W powder (calibration substance). This pattern was recorded using the following parameters: $\lambda = 1.7735$ Å, $\alpha = 22.67°$, $\delta = 50°$, exposure time: 30 s. (d): Experimental set-up for *in situ* experiments using the "$\sin^2\varphi$ method"; (e): Diffraction from {111} planes in a <111> fiber-textured film ("$\sin^2\varphi$ method"); (f): Complete Debye-Scherrer rings recorded in normal-incidence geometry used for "$\sin^2\varphi$" measurements at $\lambda = 1.5577$ Å, L = 83 mm, exposure time: 30 s.

*In situ* tensile tests were performed at the Max Planck Institute Surface Diffraction Beamline located at the synchrotron radiation source ANKA (Karlsruhe, Germany) where monochromatic X-rays, tunable in the wavelength range from 0.6 to 2.1 Å, are available. A miniature, screw-driven microtensile tester (Kammrath & Weiss GmbH, Dortmund, Germany) was mounted on the sample stage of a 2+3 circle diffractometer. Diffraction patterns were recorded using a CCD area detector, (diameter of 165 mm, 2048 x 2048

pixels). The spot size was typically 1 x 1 $mm^2$, the exposure time was 30 s, and the time required for readout and storage was 3 s per image. Two different detector positions were used for (a) characterizing the initial residual stress produced in the thin film during fabrication and for (b) monitoring the stress evolution in the film during deformation (plane stress):

(a) The equi-biaxial residual stress existing in the as-deposited state was characterized by measuring the interplanar spacing of (111)-planes exhibiting different inclination angles with respect to the film plane ($\sin^2\psi$ method). Due to the texture, strong (111)-peak intensities can be found only at inclination angles near $\psi_1 = 0°$ and $\psi_2 = 70.53°$ (Fig. 1a). In order to eliminate effects of unintentional specimen movement it was essential to detect the fcc metal (111) ring as well as at least two rings of the calibration substance (W) with a single CCD frame. The tunability of the X-ray source is crucial in this context because only by adjusting the wavelength all experimental constraints (e.g. shadowing of the tensile tester and the need for measuring the majority texture component, compare Fig. 1b) can be accounted for.

(b) For measuring the evolution of non-equibiaxial stress changes in the course of *in situ* tensile tests, the area detector was positioned “on axis” on the downstream side of the specimen, thus capable of recording the complete fcc metal (111) Debye-Scherrer ring. Elastic straining of the specimen then leads to a distortion of the shape of the ring from a circular to an elliptical one. The ellipticity of the ring is a measure of the elastic strain in the film. Further details of the methodology can be found in Ref. 11.

## 3 RESULTS

Below 300 nm film thickness cracking was observed in the Cu/Ta and Ta/Cu/Ta systems. This was first observed in the stress-strain curves, where a sudden stress drop of about half the maximum stress occurred at strains between 2 and 3%. Post mortem analysis of the samples inside a scanning electron microscope (SEM) at strains higher than 3% showed the damage morphology (Fig. 2).

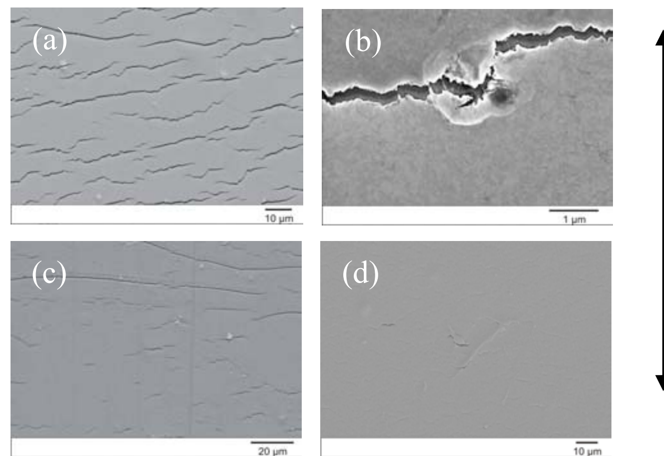


Figure 2: SEM micrographs of the surfaces of different Cu/Ta and Ta/Cu/Ta thin film systems at a total strain of 6%. The black arrow represents the direction of the external load; (a), (b) Ta/Cu/Ta sample with 100 nm Cu film thickness; (c) Cu/Ta sample with 80 nm Cu film thickness; (d) Cu/Ta sample with 640 nm film thickness.

A clear crack pattern normal to the pulling direction can be seen in Fig. 2 (a)-(c). The cracks are more easily observed in films with a Ta cap layer. No cracks could be observed in Cu/Ta systems with thicknesses larger than 300 nm (Fig. 2 (d)) and in films that did not contain any Ta at all.

## 4 DISCUSSION AND CONCLUSIONS

The stress drop observed in the stress-strain curves was correlated to crack morphology applying a model by Xia and Hutchinson [14]. The average crack distance was found to match the magnitude of the stress drop by accounting for the mismatch in the elastic moduli between substrate and metal film [13].

In this paper, we try to investigate the origin of cracking in the Cu/Ta systems. As pure Cu films did not show any cracking we assume that cracks are nucleated in the Ta layers. An additional evidence for this assumption is that Ta was deposited in the tetragonal $\beta$ phase and as such is assumed to be very brittle due to its low symmetry. The Cu film thickness then determines whether the cracks propagate through the Cu layer and cause failure or not. Following linear elastic fracture mechanics, the crack would be driven by the release of the elastic energy per unit area $W_{el}$ stored in the film:

$$W_{el} = \frac{1-\nu}{E}\sigma^2 h \quad (1)$$

where $E$ and $\nu$ are the elastic constants of the film, $\sigma$ is the stress in tensile direction and $h$ is the film thickness.

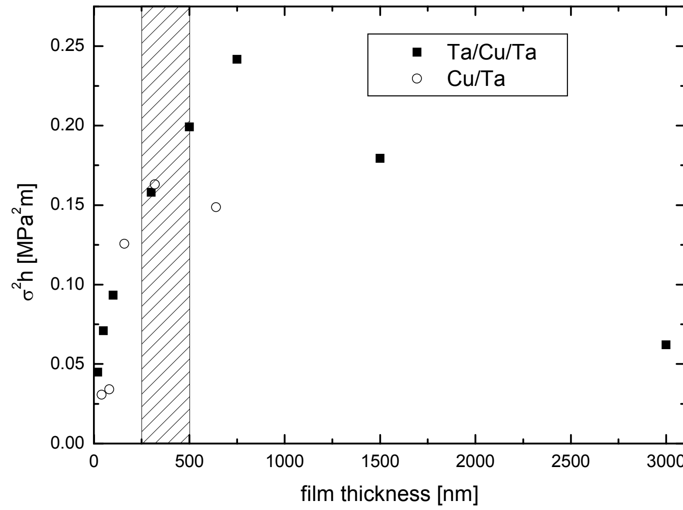


Figure 3: The fracture stress $\sigma$ squared, times the film thickness $h$, which is a measure of the elastic energy per unit area stored in the film, versus the film thickness $h$ [15]. The hatched area denotes the boundary in thickness, below which cracking has been observed.

Fig. 3 shows a maximum in elastic energy stored around the critical thickness for cracking. However, the thin films, where cracking is observed, actually exhibit a much lower elastic energy stored than the thicker films that do not crack. According to this analysis, thicker films would be expected to show cracking as well. This leads to the conclusion that not all the elastic energy stored in the thicker films can be used to propagate cracks. Other loss mechanisms such as plastic deformation at the crack tip leading to crack blunting must take place to keep the cracks from propagating into the Cu films. Further analysis which includes the effects of plasticity is required.

In summary, a size effect in the fracture behavior of Cu/Ta thin film systems has been observed. Cracks are expected to nucleate in the brittle Ta layers. The thickness dependent mechanical properties of the Cu layers then determine whether cracks propagate. Linear elastic fracture mechanics alone is not sufficient to explain the behavior observed. The thickness dependent plasticity of Cu layers needs to be included in the analysis. From a technological point of view, new *ductile* barrier materials need to be developed for electronic components on polymer substrates.

## 5 ACKNOWLEGDMENTS

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