ELASTIC PROPERTIES OF METALLIC THIN FILMS: 2D SYNCHROTRON XRD ANALYSIS DURING IN SITU TENSILE TESTING

G. Geandier,¹ P.-O. Renault,¹ S. Teat,² P. Goudeau,¹ and E. Le Bourhis¹

¹LMP-UMR 6630 CNRS, Université de Poitiers, SP2MI, Boulevard Marie et Pierre Curie, BP30179, 86962 Futuroscope Chasseneuil, France
²ALS–LBNL, 1 Cyclotron Road, Mail Stop 2-400, Berkeley, California 94720

ABSTRACT

Elastic behavior of thin films studied from in situ loading of the specimen during X-ray diffraction on a synchrotron source is presented. Model nanometric multilayer W/Au systems exhibiting different microstructures were analyzed. These films are supported by a (thin) polyimide substrate. X-ray diffraction in transmission geometry was used to study the deformations of both phases as a function of applied load. This geometry was developed with the aim of optimizing experiment time. Using 2D detectors and dynamical loading, measuring time is reduced considerably, down to a few hours compared to a one-week experiment in a laboratory. Furthermore, the in-plane strain state is measured in all directions with great precision.

INTRODUCTION

Understanding the mechanical behavior of nanostructured thin films in relation to their microstructure, in particular to the grain size, is of high importance for the development of technological applications. For a few years, there has been increasing interest in the elastic properties of thin films [1–4]. Literature data seem to show that the elastic behavior of metallic thin films can differ significantly from their bulk counterparts because of their specific microstructure (texture, defects, high density of interface, and interface mixing) and of size effects (i.e. period thickness) [5–8]. Studying and tailoring the size effect on elastic constants of polycrystalline thin films require controlling the nanostructure (grain size, residual stresses, and texture). One way to control grain size along one direction at nanometric scales is to prepare multilayers.

Elastic behavior of supported thin films can be determined by combining tensile test [9–11] or four point bending [12–14] and X-ray diffraction (XRD). XRD is phase selective and therefore a unique technique that allows determination of both the mechanical and microstructural states of the diffracting phases. This technique can be used to study how the elastic constants are affected by the period thickness independently of the multilayer components. The main disadvantage of the XRD technique is that the X-ray measurements are time consuming for thin multilayers (as the diffracting volume is quite small) even for high-flux synchrotron radiation sources.

The aim of this paper is to show how the elastic behavior of multilayers can be studied from the in situ loading of the specimen during the X-ray diffraction experiment. We report a tensile test study of W/Au multilayers films deposited on kapton substrate. As the X-ray strain
This document was presented at the Denver X-ray Conference (DXC) on Applications of X-ray Analysis.

Sponsored by the International Centre for Diffraction Data (ICDD).

This document is provided by ICDD in cooperation with the authors and presenters of the DXC for the express purpose of educating the scientific community.

*All copyrights for the document are retained by ICDD.*

Usage is restricted for the purposes of education and scientific research.

**DXC Website**  
– [www.dxcicdd.com](http://www.dxcicdd.com)

**ICDD Website**  
- [www.icdd.com](http://www.icdd.com)
measurements with relevant precision are time consuming, they were performed using synchrotron X-ray diffraction combined with a 2D detector in order to increase the amount of data which improves the analysis [15] compared to usual punctual (0D) detector analysis [16].

**SAMPLES**

The substrates used to deposit the multilayered films were 127.5 μm thick kapton dog-bone substrates; the in-plane sample dimensions were 14 × 6 mm². The substrate was cleaned with ethanol before deposition. Films were deposited by ion beam sputtering. To obtain multilayered films, W and Au were deposited sequentially and the period (thickness of W plus Au layers) was calibrated before the experiment on silicon substrate. One layer is constituted of an equal thickness of W and Au, and the gas pressure in the chamber was chosen to obtain stress free samples. The number of periods was chosen to obtain a total thickness around 700 nm. The base pressure in the growth chamber was 7.10⁻⁵ Pa, and the working pressure during film growth was approximately 10⁻² Pa. The total thickness was measured by Dektak II, a surface profilometer system, to be 640±10, 750±10, and 720±10 nm for the 12, 6, and 3 nm period films, respectively.

**EXPERIMENTAL**

Experiments were conducted on the 11.3.1 beamline at Advanced Light Source (ALS), Berkeley, California. The samples were mounted perpendicularly to the beam direction on the DEBEN™ tensile tester. Special mounting devices were used to achieve this geometry. Si powder was placed on the sample surface at the measuring point in order to correct the sample movement during loading. Energy was set at 10 keV to get the maximum number of photons and a clear signal from the two phases. The detector was set at a distance of 90 mm from the sample, to capture a major part of the Si {220} planes reflection. The geometry of the experiment is described in Figure 1.

![Figure 1. Experimental setup on the 11.3.1 beamline.](image)

Because of the films microstructure, only the strongest peaks (\{110\} for W and \{111\} for Au) will be usable during the experiments. Gold \{220\} peaks are detected on the images, but their
intensity is too low to determine the strains precisely. It is possible to increase the number of peaks for gold phase, when the sample is tilted according to the beam direction. This rotation tends to increase the intensity of the \{111\} peaks on one part of the image, but to decrease their intensity on the other part. On the same image, we have the diffracted signals from the gold sample and from the silicon powder (see Figure 2).

Transmission geometry was chosen to optimize the acquisition rate, and every image was recorded with an exposure time of 60 s. With such a short exposure time, dynamic loading of the sample was performed with loading rates ranging between 0.01 and 0.5 N/min.

The sample is loaded with a continuous rate; Figure 3 presents the evolution of the load as a function of time as recorded by the tensile module and the points where images are recorded for the 12 nm period sample. Each image is an accumulation of the signal during 60 s, so it is an average image over a load variation during 60 s. We can note that the complete experiment was finished in under 2 h, during the load increase, 34 images were recorded between 0.5 and 13 N. Other loading rates could be used depending on the sample, for example, for the sample with a rotation of 15°, a rate of 0.015 N/min was used, and 183 images were recorded between 0.5 N and 12.5 N.
STRAIN ANALYSIS

FIT2D software (http://www.esrf.eu/computing/scientific/FIT2D) is used to calibrate and reduce the data. Specific codes are used to prepare the reduced data for subsequent fitting by GNUplot software. Images are corrected for geometrical aberrations by the Brucker software and converted into tag image file format (TIFF) to be read by FIT2D. As the silicon powder signal and W/Au signals are recorded at the same time on the same frame, Si powder rings are used to calibrate the geometrical parameters of the experiment, assuming that the wavelength is stable. Sample-to-detector distance as well as image centre and detector non-orthogonality are calibrated. Using the calibrated geometry parameters (center, distance, and tilt angle) the data were divided into 1º bins ($\delta \phi$) and integrated azimuthally in a process referred to as “caking” in the FIT2D lexicon. Reduced data are saved to obtain, for each load step, classical $2\theta$ intensity diagram, one for each bin—360 diagrams per load step.

Specific code is used to generate input for GNUplot in order to fit the Si peaks that are complete over the ring and W {110} and Au {111} peaks present in the data automatically. The code can locate the peak, and extract parameters, so that the adjustment procedure in GNUplot using a Pearson VII function will start with parameters close to the final solution at the next step. Keeping $\phi$ constant, parameters from former adjustment are passed to the next load step in the GNUplot script. Parameters extracted from each peak adjustment are the position, intensity, FWHM, $M$ coefficient, and background polynomial forms (which can be of degrees 0 to 6, but linear background was used). At the end of this procedure, we have, at each loading step and for each $\phi$ positions (360 in total) all the parameters for the W, Au, and Si peaks.

Deformation in each phase (Si, W, and Au) can be determined using the following equation:

$$\varepsilon = \ln \frac{\sin \theta_0}{\sin \theta}$$  

where $\theta_0$ is the peak position at the reference load (here the first step), a measured position and not a value from theory, as we are interested in relative strains because of external loading and not residual strains.

PRELIMINARY RESULTS AND DISCUSSION

Figure 4 presents the strain evolution in the tungsten layer as a function of the applied load for the measurement made with a rotation (15º) of the sample. Because of the rotation all $\phi$ positions cannot be analyzed. Figure 5 presents the evolution of the strain in the gold layer as a function of $\phi$ positions for selected load step (right part).

Figure 4 shows that along the tensile direction (high $\phi$ positions) the layer is under tensile stress but along the transversal direction, the layer is under compressive stress (Poisson’s ratio effect). Figure 5 shows the $\phi$ positions where the layer is under tensile stresses (for $\phi$ positions close to 90º and 270º) and under compressive stresses (for $\phi$ close to 0º and 180º). At the end of the loading cycle, tungsten response presents strain saturation over a load of 10 N, probably because of the onset of plasticity in one of the two layers (probably the Au layers where similar behavior
is obtained [17]). Work is in progress to determine diffraction peak widening over plastic threshold in both phases and to confirm this hypothesis (onset in gold).

Figure 4. Strain evolution in tungsten for the 12 nm period sample (with a rotation of 15°).

For this sample (12 nm period), X-rays diffraction allows us to follow simultaneously the behaviors of Au and W phases in the multilayer film, and the peak profiles for each phase can be adjusted using standard fitting procedures without major problems. When the period decreases (for 6 and 3 nm), peaks become larger and an overlap appears as illustrated in Figure 6. For the smallest period (3 nm), we can observe only one peak close to the position of the tungsten peak, and gold peaks can be adjusted for some \( \phi \) positions.

CONCLUSION

Using a 2D detector and dynamical loading for thin films, elastic behavior study using X-ray diffraction is a good way to decrease drastically the experiment time as compared to laboratory experiments. Although an experiment in a laboratory takes one week, a similar experiment can be done on synchrotron source within a few hours, with more position in \( \phi \) and more load steps with expected improved precision. The first results show that we can easily follow the behavior of each phase in a multilayer film when the period is large enough (here over 6 nm). When the period decreases below this value, peak profiles overlapping make the analysis more and more difficult.
Figure 5. Strain evolution in gold for the 12 nm period sample (without rotation) as a function of the φ position.

Figure 6. 2θ intensity diagrams for the three samples (from top to bottom: 12, 6, and 3 nm) for different φ positions between 0° and 90°.
ACKNOWLEDGMENTS

This work has been supported partly by an ANR-05-PNANO-069 grant from the government of France and the U.S. Department of Energy who is operating ALS under Contract No. DE-AC02-05CH11231.

REFERENCES