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Abstract In-situ biaxial tensile tests within the elastic domain were conducted with W/Cu nanocomposite thin films deposited on a polyimide cruciform substrate thanks to a biaxial testing machine developed on the DiffAbs beamline at SOLEIL synchrotron. The mechanical behavior of the nanocomposite was characterized at the micro-scale and the macro-scale using simultaneously synchrotron X-ray diffraction and digital image correlation techniques. Strain analyses for equi-biaxial and non equi-biaxial loading paths have been performed. The results show that the two strain measurements match to within 1×10^{-4} in the elastic domain for strain levels less than 0.3% and for both loading paths.

Keywords: In-situ biaxial deformation; Synchrotron X-ray diffraction; Digital image correlation; Nanostructured thin films

1. Introduction

Over the recent years, great attention has been paid to miniaturized systems due to their exceptional properties and widespread applications. The continuing demands for miniaturization as well as improvement of the performance of systems extend challenges to the preparation and testing of materials with small dimensions. When stretched, freestanding metal films often experience failure at small strains typically about one percent (Huang et al., 2000; Espinosa et al., 2003; Lee at al., 2003). It has been reported that a polymer substrate could delay film failure under tensile stress, and the use of a stiff polymer substrate such as polyimide provides a stronger constraint on cracking than softer substrates (Lu et al., 2007;

Xu et al., 2010). Moreover, nanostructured thin metallic films on polymer substrates offer a combination of various mechanical and thermal characteristics that improve significantly the properties of technological devices such as sensors and stretchable microelectronics components (Li et al., 2004; Jiang et al., 2007; Xiao et al., 2008; Löher et al., 2009; Chu, 2010; Shmyreva et al., 2010; Vela-Peóa et al., 2010; Daniel et al., 2010).

In order to improve the mechanical properties of thin metallic films, it is necessary to better control their microstructure as it has been reported, for W/Cu coatings, that the deformation behavior is highly related to its microstructure (Herrmann et al., 2009). The method used herein to control the two important microstructural features in thin films, namely grain size and crystallographic texture, consists in employing a multilayer elaboration process. Introducing copper (Cu) as an immiscible interlayer for tungsten (W) thin films allows for the fabrication of nanocomposites with controlled microstructure (Valiev et al., 2002; Wang et al., 2002; Zhang et al., 2005; Girault, 2008; Gruber et al., 2009; Geandier et al., 2010a; Girault et al., 2011).

The mechanical characterization of thin films on a compliant substrate is usually achieved by different techniques such as bending tests (Jämting et al., 1997; Zhou et al., 2002; Chiang et al., 2009), nanoindentation (Baker et al., 1997; Nix et al., 1997; Wen et al., 2007; Wen et al., 2009) and tensile tests (Hommel et al., 1999; Renault et al., 2003; Böhm et al., 2004). Tensile tests are more effective and convenient to produce a uniform stress and strain fields, which enables unambiguous property measurements compared with the other techniques (Faurie et al., 2006a, 2009, 2010; Geandier et al., 2010a). Furthermore, synchrotron X-ray diffraction (XRD) provides a powerful, phase-selective and non-destructive method to probe the lattice strains in nanocomposite thin films where diffracting volumes are very small (Geandier et al., 2008; Thomas et al., 2010). Another approach for strain measurements on sample surfaces is provided by digital image correlation (DIC).

When scrutinizing the elastic behavior at small strains (less than one percent), high reliability and accuracy in strain measurements are required and can be achieved by coupling tensile tests and synchrotron XRD (Badawi et al., 2002; Renault et al., 2003; Faurie et al., 2005; 2006). Furthermore combining XRD and DIC techniques allows for the determination of the mechanical behavior both at the micro- and macro-scales. In the case of elastically anisotropic materials, the combination of these two techniques can be very useful to better interpret the complex mechanical response, and to study the inter-grains interactions that arise in polycrystalline thin films due to the elastic anisotropic behavior of grains (Faurie et al., 2009). The commonly used method, i.e., uniaxial tensile tests, is not describing the realistic loading conditions of materials in-service contrary to biaxial tensile tests, which allow the exploration of all possible plane stress states. A biaxial testing machine is now available and

optimized at DiffAbs beamline of the French synchrotron radiation facility (SOLEIL, Saint-Aubin). In particular, equi-biaxial and non-equi-biaxial loadings (different longitudinal and transverse loadings) can be applied to thin films deposited on flexible substrates (Djaziri et al., 2010; Geandier et al., 2010a).

In this work, synchrotron XRD and DIC are coupled with a biaxial testing device to measure with high accuracy elastic strains of W/Cu nanocomposite thin films deposited on a polyimide substrate. The paper is organized as follows. Section 2 describes the experimental method; the sample and the testing device are presented. This section also provides details on the in-situ biaxial tensile tests performed on W/Cu composite. Section 3 gives the results of measured strains by XRD and DIC that are compared to each other for different two loading paths. The results are discussed in terms of performance and purpose of each used technique. Section 4 focuses on conclusions.

2. Experimental details

2.1. Sample preparation and biaxial tensile device

W/Cu composite thin films were deposited on 127-µm-thick polyimide (Kapton®) cruciform substrate 200 and 650 µm-thick (001) oriented Si wafers, and were produced at room temperature by physical vapor deposition (PVD) with an Ar^+-ion-gun sputtering beam at 1.2 keV (Kaufman ion source) in a NORDIKO, Hampshire-3000 system. The base pressure of the deposition chamber was 7×10^{-5} Pa, while the working pressure during film growth was approximately 10^{-2} Pa. The films were fabricated with 60 periods of W/Cu, and the deposit time of each W and Cu layers was 60 s and 7 s, respectively. Average W and Cu growing rates were previously calibrated by X-ray reflectivity (XRR) for thick layers (between 50 and 100 nm) and were found to be 0.05 nm s^{-1} and 0.07 nm s^{-1}, respectively. The expected thicknesses for both W and Cu sub-layers are 3 and 1 nm, respectively. The X-ray reflectivity was measured using a Seifert XRD 3003 diffractometer operating with a Cu X-ray source (linear focus, $\lambda = 0.15406$ nm). The total film thickness was controlled using a Dektak II-a surface profilometer system and crystallographic texture and residual stress analyses were carried out using the same diffractometer (Seifert XRD 3003 diffractometer with a punctual focus, $\lambda = 0.15418$ nm). The above presented analyses have been carried out on the sample deposited on (001) Si wafers.

The sample deposited onto Kapton® has been loaded biaxially thanks to the biaxial testing device developed on the DiffAbs beamline at SOLEIL synchrotron. This machine was designed to allow for loadings along two perpendicular axes of cruciform substrates coated at their center only by the studied films (no adhesion layer was deposited). Further information about this biaxial device can be found in (Geandier et al., 2010a). The polyimide substrate

(Kapton® HN from DuPont™) is expected to behave elastically in the investigated strain range (maximum of 0.4 %). Fig. 1 shows the machine with a gripped cruciform specimen installed in the DiffAbs diffractometer.

2.2. Strain measurements

Strain measurements have been performed for an equi-biaxial loading and a non equi-biaxial loading using both X-ray diffraction and digital image correlation techniques. Therefore, the mechanical behavior of the sample was obtained at two different scales. At the micro-scale, synchrotron X-ray diffraction was used to measure strains within the thin metal film over coherent diffraction domains. At the macro-scale, digital image correlation was used to assess the strain in the polyimide cruciform substrate. The image of the bottom side of the substrate (the uncoated side) was captured with the CCD camera (see Section 2.2.1).

An initial load has to be applied for the installation of the specimen into the biaxial tensile device in order to avoid sample drift during the biaxial tensile test (Geandier et al., 2008). In the following, the applied loads are incrementally increased from the initial loading state, and range between 0 and 40 N (Tables 1 and 2).

2.2.1. Digital image correlation

Digital image correlation (DIC) is an optical technique to measure displacement and strain fields on an object surface by capturing images of the object surface at different states. One state is recorded before loading, i.e. the reference image, and the other states are subsequent images of the deformed object. DIC uses random patterns of gray levels of the sample surface to measure the displacement via a correlation of a pair of digital images.

In this study, the specimen rear surface was spray-painted with a speckle pattern to generate a contrast in the uniform specimen face (Fig. 2), and white light illumination was used to obtain black and white (B/W) images. Digital images of the specimen surface were captured with an optical camera fixed under the testing device (Fig. 1). The used optical camera is composed of a telecentric lens (× 1.0 from Edmund Optics, York, UK) and a CCD camera (Pixelfly from PCO AG, Kelheim, Germany, QE-12 bit dynamic range, 1392×1024 pixel definition, and a pixel size of 6.45 × 6.45 μm², black and white). The size of the region of interest at the sample surface is 9 × 6.3 mm².

For each applied load, the specimen was let relax for about 10 minutes (see Section 3.2). Then, a set of 10 images of the rear surface of the specimen was recorded before and after the XRD measurements. Image correlation and subsequent strain calculations were achieved using a Q4-DIC technique in which the displacements are assumed to be described by Q4P1-shape functions relevant to finite element simulations. The region of interest (ROI) of the sample surface is thus discretized with continuous 4-noded (Q4) elements (Besnard et al.,

2006). The Q4-DIC procedure is implemented in a MATLAB^{TM} code, and is used in the present work to measure the displacement and strain fields of the hybrid composite under biaxial loadings.

2.2.2. Synchrotron X-ray diffraction

Because of small diffracting volumes and the complex microstructure of the composite (mainly the nanometric crystallite size), *in situ* characterization by standard X-ray sources is time consuming, and more often does not allow for the measurement of strain distributions accurately. Synchrotron X-ray diffraction represents a powerful and non-destructive method to provide high accurate information with short acquisition times thanks to the high flux and brightness of synchrotron radiation sources.

The X-ray energy and beam size have been set to around 8.8 keV and 0.320 × 0.370 mm^2 (FWHM, H × V) respectively. We checked by finite element analysis (FEA) that for an equi-biaxial loading, uniform strains were generated in a central area 8 mm in diameter, which is larger than the irradiated area. For a non-equi-biaxial loading, the size of the uniform strain area is depending on the load F_x / F_y ratio (F_x and F_y being the applied forces along x and y axes, respectively, see Fig. 1). However, it is still larger than the employed X-ray beam size (Geandier et al., 2010a). The measurements have been performed for different {hkl} lattice planes, at different tilts (17 Ψ angles) and different azimuthal angles (two Φ angles) using a two-dimensional detector, with the XPAD detector developed by SOLEIL (Medjoubi et al., 2010). It consists of eight modules of seven hybrid integrated circuits (chips) on a single silicon sensor. Each of these chips consists of 80 × 120 pixels measuring 130 µm along each side, and the complete imager has a footprint of 7.28 × 12.48 cm^2. This type of hybrid-pixel detector is very attractive for its fast read-out time, high dynamic range and signal-to-noise ratio. The use of this two-dimensional detector offers relatively reliable X-ray strain measurements with very short counting times comparing to punctual detector which has been used for preliminary X-ray diffraction characterization (Djaziri et al., 2010). During latter experiments, a similar sample was studied under equi-biaxial loadings to check the strain homogeneity predicted by finite element analysis. Typically, each diffraction pattern has been recorded during about 30 s. Diffraction pattern represents a small part of Debye-Scherrer ring (inset of Fig. 3) because of the large specimen to detector distance (536 mm). Each 2D diffraction pattern was transformed, after calibration and correction, to a classical 1D diffraction peak thanks to a radial integration (Fig. 3). The obtained diffraction peaks were then fitted by a Pearson VII function considering a linear background. The in-grain strain was obtained by detecting the diffraction peak position shift for each loading state. Therefore, the lattice strain can be determined independently of the residual stress in the material. The lattice

strain $\{\varepsilon\}_{\Phi\Psi}^{hkl}$ corresponding to {hkl} diffracting planes and a scattering vector with orientation (Φ, Ψ) is given by:

$$\{\varepsilon\}_{\Phi\Psi}^{hkl} = \ln\left(\frac{d_{\Phi\Psi}}{d_{\Phi\Psi}^{(0)}}\right) = \ln\left(\frac{\sin\theta_{\Phi\Psi}^{(0)}}{\sin\theta_{\Phi\Psi}}\right) \approx \frac{d_{\Phi\Psi} - d_{\Phi\Psi}^{(0)}}{d_{\Phi\Psi}^{(0)}} \quad (1)$$

where $d_{\Phi\Psi}^{(0)}$ is the reference lattice spacing, and $\theta_{\Phi\Psi}^{(0)}$ the associated reference diffraction angle. $d_{\Phi\Psi}$ and $\theta_{\Phi\Psi}$ are the lattice spacing and the diffraction angle, respectively, for the loaded states. In-grain elastic strains measured by XRD depend on {hkl} plane orientation defined by the in-plane azimuthal Φ and tilt Ψ angles. The conditions of the experimental measurements were quasi-static, namely, measurements along the two azimuthal angles $\Phi = 0°$ and $\Phi = 90°$ were performed independently.

3. Results and discussion

3.1. Structure of tungsten phase

Tungsten and copper were alternatively sputtered to fabricate films with a total thickness evaluated to be about 217 ± 10 nm. The stratification could be detected using XRR and the average period thickness of the W/Cu multilayer has been determined to be 3.7 nm (Fig. 4). Phase structure of W sub-layers is body centered cubic (α–bcc). The crystallographic texture analysis showed that W sublayers exhibit a strong α-{110}-fiber texture (Fig. 5). The texture is not perfectly sharp and shows a degree of scatter which can be quantified by the full width at half maximum (FWHM) of the rocking curve peaks. It was determined to be approximately 18° on the {110} planes family. In a former study, transmission electron microscopy (TEM) analyses have revealed that the sub-layer grains have equiaxed morphology with an average size close to the sub-layer thickness (Girault, 2008; Girault et al., 2011). Due to the small grain size, the observed diffraction peaks are very large (Fig. 3). The W/Cu thin films are subjected to high compressive residual stresses. Global residual stresses of the film, measured by the curvature technique were found to be -1.5 GPa while residual stresses within W grains, obtained by X-ray stress analysis (XSA) were found equal to -3.5 GPa (Girault, 2008). Table 3 summarizes the different characteristics of the studied composite.

3.2. Digital image correlation strains

As presented previously, a Q4-DIC technique allowed for the measurement of the displacement and strain fields of the sample surface (non coated side). It is based on the comparison of digital images taken at different loading states in which the analyzed region or the region of interest (ROI) of the sample surface is subdivided in several zones of interest (ZOI) which constitute a mesh of continuous Q4-finite elements (square elements).

The displacement field is obtained by comparing and correlating the gray-scale distribution from one image to the other. The basic equation of the DIC method is related to the conservation of gray levels:

$$g(\mathbf{x}) = f(\mathbf{x} + \mathbf{u}(\mathbf{x})) \tag{2}$$

where f(**x**) and g(**x**) are gray level functions corresponding to the reference and deformed configurations at each pixel point of coordinate **x**. **u**(**x**) is the displacement function in the measured direction. The global gray level conservation is written by minimizing the following functional (Besnard et al., 2006):

$$\eta^2 = \iint_{ROI} \Phi^2(\mathbf{x})\, d\mathbf{x} \tag{3}$$

where the local correlation residual, Φ(**x**), is given by

$$\Phi(\mathbf{x}) = \left| f(\mathbf{x} + \mathbf{u}(\mathbf{x})) - g(\mathbf{x}) \right| \tag{4}$$

The minimization of the function is done over each ZOI by searching a bilinear form of the displacement field. When the optimal parameters of equation (2) for every ZOI are found, the corresponding displacement components of each point in the deformed and the reference ZOI can be obtained. Thus, the displacement gradient can be determined. Considering small strains (< 0.3%) and very small rotations, the infinitesimal strain tensor can be expressed in terms of the displacement field:

$$\varepsilon_{ij} = \frac{1}{2}\left(\frac{\partial u_i}{\partial j} + \frac{\partial u_j}{\partial i} \right) \tag{5}$$

where (i,j) = (x,y), and u_i and u_j are displacement components.

The accuracy of the measurements depends mainly on the quality of the texture which will affect the selection of the ZOI size. The texture of the sample surface is shown in Fig. 2. Random dots were obtained by spraying a white paint prior to the experiment. A priori analyses of the image texture have been performed to evaluate the performance of the Q4-DIC technique. A 0.5-pixel displacement is artificially applied to create a 'deformed' picture (Besnard et al., 2006). It is shown that the displacement uncertainties decrease as the ZOI size becomes greater than 16 pixels. 32-pixel elements have been chosen as a good compromise between displacement uncertainty and spatial resolution. The corresponding standard displacement uncertainty is about 10^{-2} pixel. This uncertainty level would be presumably lower if the texture had a larger dynamic range. In the present case, only a very small part (800 gray levels) of the dynamic range of the CCD camera (12 bits) was used. However, the strain results show a good performance of the Q4-DIC technique with a strain resolution of 10^{-4} (strain measurement uncertainties are based on two standard deviations).

Typical in-plane displacement fields (u_x and u_y components) are shown in Fig. 2. The two displacements fields are similar in both directions. The iso-displacements lines are parallel and equi-distant revealing the uniformity of the displacement field. The similar distances between the iso-displacements lines in the two directions is the signature of an equi-biaxial loading. In Fig. 6, main strains calculated by Q4-DIC as functions of the applied force are presented for equi-biaxial loadings. First, the strain of the composite polyimide/thin film versus applied load is linear over the load range investigated herein. Second, the two components of the in-plane strains are slightly different, and the difference increases with the applied force. It mainly results from load states that are not exactly equi-biaxial. Third, for each component of the in-plane strains, the two curves corresponding to measurements carried out before and after X-ray measurements exhibit no significant deviations (Fig. 6). This result clearly demonstrates that polyimide substrate relaxation is negligible during X-ray measurements. This is to be kept in mind on analyzing diffraction data detailed in the next section. The evaluated in-plane strains for a load of ~37 N (the equivalent stress value applied to the branches is ~13 MPa) are as follows: $\bar{\varepsilon}_{xx} = 0.20\%$ and $\bar{\varepsilon}_{yy} = 0.21\%$.

To look at strain homogeneity, different circular ROI were set on the images recorded during the tensile tests. Regions of 1, 2, 3 and 4 mm in radius were used and the obtained results have been compared to each other. Noticeably, the measured strains for 1mm-ROI show the highest strain. The other ROI present average strains very close to that evaluated from the full image. Nevertheless, the relative deviation of the 1mm-ROI strain from the other ROI is of only 3%. This deviation should be due to the lack of statistic on the 1mm-ROI. In conclusion, the DIC technique depicts homogeneity of strains on the acquired images for the equi-biaxial loading as well as for the non-equi-biaxial loading.

3.3. Synchrotron X-ray diffraction strains

The lattice strain is obtained by measuring the Bragg-peak position shift with respect to its position in the unloaded state. A 2θ position shift of the {211} Bragg peak of tungsten phase under loading is observed in Fig. 3. It is worth noting that due to small applied strains, the displacements of diffraction peaks are very small. Besides, the small grain size of the investigated material makes the diffraction peaks large. The use of a 2D detector taking advantage of the high flux and brilliance of the synchrotron radiation allows for improved measurements by saving counting times and avoiding substrate and metallic thin film relaxation. The used XPAD 2D detector is a photon counting device with high dynamic range, high counting rate, and short read-out time compared to CCD detectors (Medjoubi et al., 2010). A typical 2D diffraction pattern is presented in the inset of Fig. 3. The analysis, as mentioned in Section 2.2.2, consists in making radial integration of the 2D pattern to obtain

the classical diffraction peak. Assuming plane-stress conditions, the strain equation is written for a biaxial loading:

$$\{\varepsilon\}_{\Phi\Psi}^{hkl} = \frac{1}{2} S_2^{hkl}\, \overline{\sigma}_{\Phi}\, \sin^2\Psi + S_1^{hkl}\left(\overline{\sigma}_{xx} + \overline{\sigma}_{yy}\right) \quad (6)$$

where $\overline{\sigma}_{\Phi} = \overline{\sigma}_{xx}\cos^2\Phi + \overline{\sigma}_{xy}\sin 2\Phi + \overline{\sigma}_{yy}\sin^2\Phi$; $\overline{\sigma}_{xx}$ and $\overline{\sigma}_{yy}$ are the macroscopic stresses applied to the thin film.

3.3.1. Equi-biaxial strains

When the applied loadings are equi-biaxial ($\overline{\sigma}_{xx} = \overline{\sigma}_{yy} = \overline{\sigma}$, and $\overline{\sigma}_{xy}$ =0), the strain equation reduces to:

$$\{\varepsilon\}_{\Phi\Psi}^{hkl} = \frac{1}{2} S_2^{hkl}\, \overline{\sigma}\, \sin^2\Psi + 2 S_1^{hkl}\overline{\sigma} \quad (7)$$

Since tungsten is a locally isotropic material, all the mechanical models lead to the same values for X-ray elastic constants (XEC) $S_1^{hkl} = -\frac{\nu}{E}$ and $\frac{1}{2}S_2^{hkl} = \frac{1+\nu}{E}$, where E is the Young's modulus and ν is the Poisson's ratio of the material (Hauk, 1997; Welzel et al., 2005; Faurie et al., 2009).

Fig. 7 and 8 show the plots of the measured X-ray $\{\varepsilon\}_{0°\Psi}^{211}$ and $\{\varepsilon\}_{90°\Psi}^{211}$ strains within the tungsten phase as a function of $\sin^2\Psi$ for two azimuthal angles $\Phi = 0°$ and $\Phi = 90°$, respectively, for equi-biaxial loadings. The plots clearly show a linear relationship between strains and $\sin^2\psi$ over a large number of Ψ angles (17 angles ranging from 0 up to 70°) at $\Phi = 0°$ and $\Phi = 90°$, which allows reliable strain measurements. This behaviour is characteristic of macroscopically elastically isotropic materials i.e., quasi isotropic materials where surface effects induced anisotropy (Faurie et al., 2006b; Welzel et al., 2009) can be neglected, It is worth noting that the studied specimen is composed of 60 W sublayers of 3.7 nm thickness each. The $\varepsilon - \sin^2\Psi$ lines are expected to cross at one point $\sin^2\Psi^* = \frac{2\nu}{1+\nu}$, which corresponds to a strain equal to zero whatever the applied stress value (equation (7)). Noticeably, all the linear fits meet in a very small strain range from -10^{-5} to 10^{-4} emphasizing the reliability of the measurements.

In Fig. 9, the results of the in-plane strains measured by X-ray diffraction as a function of the applied load are presented. The two strain components ε_{xx} and ε_{yy} are equal to the sum of the slope and the intercept of $\varepsilon - \sin^2\Psi$ curves at $\Phi = 90°$, and $\Phi = 0°$, respectively. The strain evolutions are linear as expected for loadings within the elastic domain. This figure shows that the two strain components are slightly different. This difference is increasing with the

applied force as already observed for DIC strain measurements. Moreover, DIC and XRD strains superimposed within experimental uncertainty for each component. Noticeably, the difference between ε_{xx} and ε_{yy} is significant and equivalent for both techniques. The X-ray strains for ~37 N are as follows: $\varepsilon_{xx} = 0.19$ % and $\varepsilon_{yy} = 0.21$ %. Poisson's ratio can also be extracted from the slope of these equations:

$$\varepsilon_{zz} = \left(\frac{2\nu}{\nu - 1}\right) \times \varepsilon_{xx} \text{ and } \varepsilon_{zz} = \left(\frac{2\nu}{\nu - 1}\right) \times \varepsilon_{yy} \quad (8)$$

These two quantities must be equal in the case of a perfect equi-biaxial loading, which is not the case in the present work (Fig. 9) as already discussed. Strain ε_{zz} corresponds to the out-of-plane component, which is equal to the intercept of the ε - $\sin^2\Psi$ curves. We found a value of $\nu = 0.274 \pm 0.010$, which is slightly less than Poisson's ratio of bulk W (0.284; Smithells, 1976) and close to that obtained from the intersection point at $\sin^2\Psi^* \approx 0.43$.Poisson's ratio measured for a W single layer is ranging from 0.260 to 0.284 (Geandier et al., 2010a).

3.3.2. Non-equi-biaxial strains

For a non-equi-biaxial loading, the measured strains within the tungsten phase, for $\Phi = 0°$ and $\Phi = 90°$, respectively, are written as (Hauk, 1997):

$$\{\varepsilon\}_{0,\Psi} = \frac{1}{2} S_2^{hkl} \, \overline{\sigma}_{xx} \sin^2\Psi + S_1^{hkl} \left(\overline{\sigma}_{xx} + \overline{\sigma}_{yy}\right) \quad (9)$$

$$\{\varepsilon\}_{90,\Psi} = \frac{1}{2} S_2^{hkl} \, \overline{\sigma}_{yy} \sin^2\Psi + S_1^{hkl} \left(\overline{\sigma}_{xx} + \overline{\sigma}_{yy}\right) \quad (10)$$

The evolutions of the two in-plane strain components as a function of applied couple force for the non equi-biaxial path are plotted in Fig. 10. The loading path encompasses three almost equi-biaxial loadings. In a first step, F_x was increased up to 20 N while F_y was maintained constant. In a second step, F_y was increased up to 38 N and F_x was maintained at 20 N. In a third and last step, F_x was increased up to 38 N and F_y was maintained constant. The increase of the load in only one branch induces an increase of the strain in this branch and a decrease of the strain in the other perpendicular branch due to Poisson's effect as observed. DIC and XRD strains superimpose within experimental uncertainty (10^{-4}) for each component all along the complex loading path.

The evolution of ε_{zz} as a function of the sum of ε_{xx} and ε_{yy} is linear:

$$\varepsilon_{zz} = \left(\frac{\nu}{\nu - 1}\right) \times \left(\varepsilon_{xx} + \varepsilon_{yy}\right) \quad (11)$$

From the slope of this equation, Poisson's ratio is extracted and it is found to be about 0.286 ± 0.010. This value is larger than that obtained for an equi-biaxial loading, but is still in the expected range and very close to that of bulk W.

3.4. Accuracy of DIC and XRD strains

The two approaches namely DIC and XRD investigate the behaviour of material at the macro-scale and the micro-scale for non-destructive and non-contacting direct strain measurements. A comparison of the two approaches for measuring in-situ plane strains shows that the strain measurements are of good accuracy and adaptability for various stress conditions. The strains by DIC and XRD techniques are equal within 1×10^{-4} for both loading paths. Through these tests, we can assure that the combined synchrotron X-ray diffraction and image correlation analyses could be used with efficiency and reliability in biaxial tensile tests of thin films deposited on polymer substrate. The consequence of the accuracy of the two strain measurements techniques is that the applied strain is determined to be transmitted unchanged in the elastic domain through the metallic film - polymeric substrate interface although no adhesion layer was used as already reported for gold films in Geandier et al. (2010b).

4. Conclusion and perspective

In this study, DIC and XRD techniques were utilized to investigate, at two different length scales, the elastic behavior of nanostructured thin films under two different biaxial loading conditions. The complete strain tensors of the studied sample were measured. Both methods, XRD and DIC, are non-destructive and non-contacting for direct strain measurements with good accuracy. Synchrotron XRD is a powerful and useful technique to measure elastic strains of polycrystalline materials especially for small sized samples such as thin films. It is worth noting that 2D detector allows for fast data acquisition times, and thus 'dynamic' in situ investigations. DIC is used to track the mechanical response of the substrate. The transmission of strains at the film-substrate interface was demonstrated, and the good adhesion of the thin film to the substrate at small applied strains was proven. DIC results showed that after a loading period of 10 minutes, polyimide substrate relaxation can be neglected in the applied loading range (i.e., up to 37 N corresponding to 13 MPa mean stress at substrate center). Uncertainties of the order of 10^{-4} were determined for both DIC measurements and synchrotron XRD measurements, respectively. The results show the two strain measurements match to within 1×10^{-4} in the elastic domain for strain levels less than 0.3%, and for both loading paths.

Moreover, it is to be highlighted that the studied W thin films presented a single α-{110}-fiber texture component thanks to the control of the microstructure by introducing a given amount of Cu as an immiscible interlayer. The crystallographic texture in W thin films allows

for a simple and straightforward analysis of the mechanical properties. However, for elastically anisotropic polycrystalline films (Dong et al., 1998; Martinschitz et al., 2009; Yokoyama et al., 2009; Faurie et al., 2010), micromechanical models taking account of the actual microstructure have to be used due to the interaction of differently oriented grains. For instance, polycrystalline thin films with mixed crystallographic textures can be subjected to heterogeneous triaxial stress states arising from grain interactions that are crucial to the understanding of the mechanical behavior of these films (Nix, 1989; Vinci, 2008; Vodnick et al., 2010). Conversely, thin films with perfect fiber texture have uniform biaxial stress states and present enhanced properties such as improved stability, electromigration reliability and high strain hardening (Tomar et al., 2008; Vodnick et al., 2010).

The proposed method of combining the two techniques makes it possible to carry out dynamic tests and tackle the material mechanical behaviour at two scales simultaneously. The method can deal with high strains including plastic deformation during the biaxial tensile tests.

Figure 1

Biaxial testing machine on DiffAbs diffractometer at SOLEIL synchrotron. The optical camera is located under the machine. A two-dimensional (XPAD) detector was used for the X-ray diffraction experiments. The X-ray beam size is 320 × 370 μm^2 (FWHM, H × V). The sample-to- detector distance is about 53.6 cm. The inset shows the geometry of the diffraction set up used for the cruciform samples. The biaxial tensile stresses are applied along **x** and **y** axes. The direction of the scattering vector **q** is defined by the azimuthal angle Φ from **x** and the polar angle Ψ from the sample normal direction **z**.

Figure 2

(Color online) Visualization of the acquired image with displacements fields along a) vertical and b) horizontal directions corresponding to displacement components u_x and u_y respectively for the equi-biaxial loading T9 ~ 37 N. The specimen rear surface of Kapton substrate has been spray-painted with a speckle pattern. A white light emitted diodes (LED) illumination was used to obtain B / W images. The size of the region of interest (ROI) is 9 × 6.3 mm^2.

Figure 3

W {211} Bragg peaks measured for the unloaded (T0 = 0 N) and loaded (T9 = 37 N) states (Ψ = 29.3°, Φ = 0°). Discontinuities result from the discrete structure of the XPAD detector. The inset shows a 2D diffraction pattern after image correction.

Figure 4

X-ray reflectivity curve of W/Cu composite (60 periods).

Figure 5

Ψ scans obtained for the {110} planes family of tungsten phase for W-Cu thin films deposited on polyimide substrate. A comparison is made with a monolithic W film deposited under the same conditions (studied in Geandier et al., 2010a). Coexistence of two fiber crystallographic texture in W films is revealed by sharp peaks at Ψ =0° for {110} - fiber texture and at Ψ =35° (the correspondent symmetric peak at Ψ =-35°) for {111} - fiber texture. In contrast, W-Cu films present a {110} - fiber texture which is characterized by sharp peaks at Ψ =0° and at Ψ =60° (the correspondent symmetric peak at Ψ =-60°).

Figure 6

Stains measured by DIC (before and after XRD measurements) as functions of the applied load for the equi-biaxial path. $\overline{\varepsilon}_{xx}$ (circular symbols) and $\overline{\varepsilon}_{yy}$ (squared symbols) are the two in-plane strain components. Solid lines are obtained by linear fit of the experimental data. The dark lines represent fits of $\overline{\varepsilon}_{xx}$ data, and gray lines fits of $\overline{\varepsilon}_{yy}$ data.

Figure 7

X-ray strains within W grains as functions of $\sin^2\Psi$ for the (211) reflection at azimuthal angle $\Phi = 0°$ for the equi-biaxial loading. Solid lines are linear fits of the experimental data.

Figure 8

X-ray strains within W grains as a function of $\sin^2\Psi$ for the (211) reflection at azimuthal angle $\Phi = 90°$ for the equi-biaxial loading. Solid lines are linear fits of the experimental data.

Figure 9

Superposition of X-ray elastic strains (ε_{xx} and ε_{yy}) and DIC strains ($\overline{\varepsilon}_{xx}$ and $\overline{\varepsilon}_{yy}$) as functions of the applied load (equi-biaxial loading). Strains components $\overline{\varepsilon}_{xx}$ and $\overline{\varepsilon}_{yy}$ correspond here to the average values of strains measured before and after XRD measurements by DIC. Open symbols represent experimental data from DIC measurements, and solid symbols correspond to experimental data from XRD measurements. Solid lines are linear fits of the experimental data. The vertical error bars represent the uncertainty of XRD and DIC strain measurements (1×10^{-4}).

Figure 10

Superposition of X-ray elastic strains (ε_{xx} and ε_{yy}) and DIC strains ($\overline{\varepsilon}_{xx}$ and $\overline{\varepsilon}_{yy}$) as functions of the applied load (equi-biaxial and non equi-biaxial loadings). Strains components $\overline{\varepsilon}_{xx}$ and $\overline{\varepsilon}_{yy}$ correspond to the average values of strains measured by DIC before and after XRD measurements. Open symbols represent experimental data from DIC measurements, and solid symbols correspond to experimental data from XRD measurements. The vertical bars represent the uncertainty of XRD and DIC strain measurements.

Table 1 Applied loads for the equi-biaxial test before and after XRD measurements.

Loading step	Applied load before XRD (N)	Applied load after XRD (N)
T1	3.20	3.05
T2	10.50	10.15
T3	17.35	16.90
T4	20.65	20.30
T5	24.00	23.60
T6	27.40	26.95
T7	30.90	30.25
T8	34.25	33.60
T9	37.45	36.90

Table 2 Applied loads for the non equi-biaxial test (i.e., average values of levels after and before XRD measurements).

	F_X (N)	F_Y (N)
T1	6	0
T2	16	0
T3	20	0
T4	20	6
T5	20	16
T6	20	20
T7	20	26
T8	20	34
T9	20	38
T10	34	38
T11	38	38

Table 3 Characteristic features of the studied W/Cu composites deposited on Kapton® substrates.

Thin film	Substrate	Total thin film thickness (nm)	Crystallographic texture of W	Residual stresses in W (GPa)	Global residual stresses (GPa)
W/Cu	Kapton® HN	217	{110}	-3.5	-1.5

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