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ESRF

Experiment Report Form

<table>
<tr><td>ESRF</td><td>Experiment title:
Elasto-plastic behaviour of nanometric metallic multilayers</td><td>Experiment number:
02-02-671</td></tr>
<tr><td>Beamline:
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<tr><td>Shifts:
12</td><td>Local contact(s):
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Report:

Our objective was to study the size effect on the mechanical behaviour of W and Cu layers when the thickness is reduced down to a few tenths of nanometres. We were interested in both the elastic coefficients and the appearance of plastic yield. The system chosen here was a series of three W/Cu multilayers with a period ranging from 24 down to 6 nm and a constant tungsten over copper thickness ratio (1/3): W6/Cu18 nm, W3/Cu9 nm, and W1.5/Cu4.5 nm. These metallic multilayers were deposited by magnetron sputtering on 125 µm thick Kapton® dogbone substrates for tensile tests with a total film thickness of about 1 µm.

We performed in situ tensile tests thanks to a dedicated Deben™ deformation device mounted on the 7C diffractometer at BM02 beam line, with a constant X-ray energy of 8.95 keV and a 0D detector. We scheduled the 12 allocated shifts as following:

- 2 for energy and focalisation adjustment, and preliminary tests on our samples;
- 4 for measurements on the W6/Cu18 specimen;
- 6 for measurements on the two other specimens (W1.5/Cu4.5 and W3/Cu9).

8 or 9 increasing loads were applied to each sample and $\theta/2\theta$ X-ray measurements were done with several ψ orientations and two ϕ values (0 and 90°) for each loading level. Since the tungsten and copper layers are <110> and <111> fibre-textured respectively, the chosen ψ values correspond to pole directions. The whole load range was supposed to be included in the elastic strain domain of the multilayers (according to prior laboratory tests); we lacked time to study the plastic domain and even to check that the strain remained purely elastic by performing X-ray measurements while unloading the sample. It should be noted that these measurements would not have been possible with a conventional X-ray source, even with very long

acquisition times, since uncertainties are of the same order as the peak shifts; here the X-ray synchrotron source allowed us to perform good quality measurements, with a rather high dynamic, in a quite short recording time (between 2 and 4 hours for each loading step).

This preliminary report is mainly based on the analysis of the W(310) and Cu(222) diffraction peaks (for all the ψ values) for the three specimens. The other peak positions in the investigated 2θ range are more difficult to determine with good precision since W and Cu peaks overlap. Moreover, as layer thickness decreases, peak broadening makes peak deconvolution more difficult. Figures 1 and 2 show $\theta/2\theta$ diagrams performed for the 6N loading state on the W6/Cu18 multilayer for several ψ values and on each multilayer for a given ψ value, respectively.

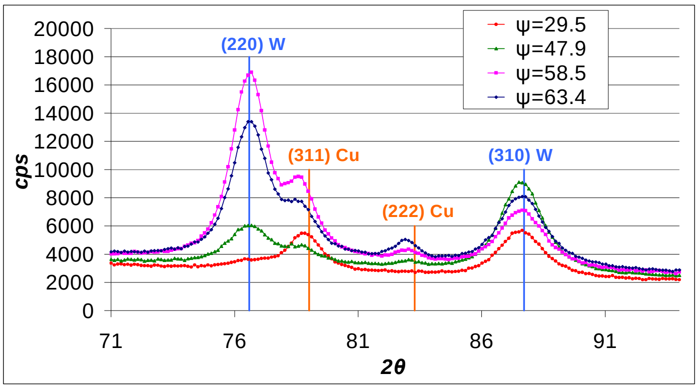

Figure 1: Diffraction diagrams of the W6/Cu18 multilayer for different ψ–values but a constant applied load of 6N (λ_{XR} = 0.1387 nm).

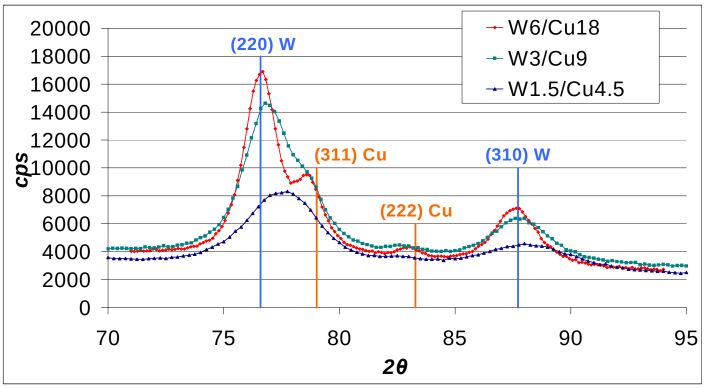

Figure 2: Diffraction peak form for ψ=58.5° for each multilayer at a constant load of 6N: peak broadening increases as the period decreases (λ_{XR} = 0.1387 nm).

The W(310) peak shifts as a function of the applied force are shown in figure 3 for four ψ values. These measurements allow calculating the strain associated to each loading increment (the first loading state T0 is taken as a reference). For (310) plane orientation close to ψ = 30°, the 2θ peak position is almost constant whatever the applied force, which implies that the elastic strain remains nil. For ψ>30°, the peak position is decreasing when the force increases, which corresponds to a dilatation of lattice planes. Furthermore, the four curves present a common intersection point with abscissa around 6.5 N: here the applied tensile stress is balancing the residual compressive stress in tungsten layers. Considering in figure 4 the XRD-strain in tungsten as a function of the applied force for the 3 periods at ψ=63.4°, we can already suppose a size effect since linear regression slopes are different for each period. Obviously, the (ε,F) slope increases when the multilayer period decreases, especially for the W1.5/Cu4.5 sample. An elastic softening of W layers and a possible interface roughness may contribute to this variation.

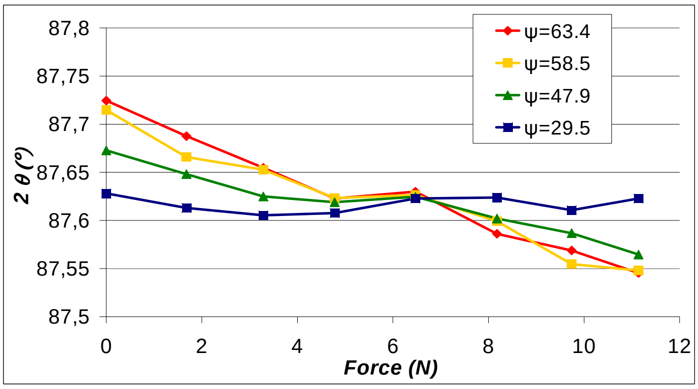

Figure 3: Evolution of the W(310) diffraction peak position as a function of the applied load for 4 values of the ψ angle, in the W6/Cu18 multilayer (λ= 0.1387 nm).

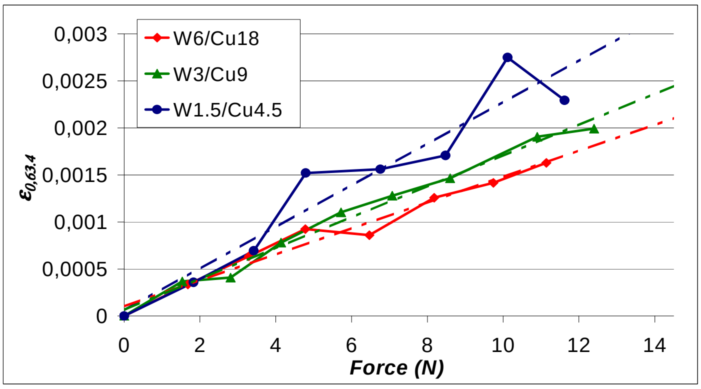

Figure 4: Strains deduced from the shift of the W(310) diffraction peak as a function of the applied load, for ψ=63.4°, for the 3 multilayers.

Taking into account all $\varepsilon_{0,\psi}$ measurements, we can plot $ln(a_\psi)$ versus $sin^2\psi$ curves, a_ψ being the lattice parameter in the ψ direction. In the case of tungsten, a linear behaviour is observed as shown on figure 5 for the W6/Cu18 sample. The slope of the linear regressions increases regularly with the applied load. The

longitudinal applied strain ε_{11} can finally be extracted from the slope and intercept of each curve, taking “T0” as a reference. The results obtained for the three specimens are presented in figure 6. These plots confirm the size effect previously mentioned (Fig.4): the slope of the (ε,F) curve is for the W1.5/Cu4.5 multilayer than for the other ones, which clearly indicates a softening of the material.

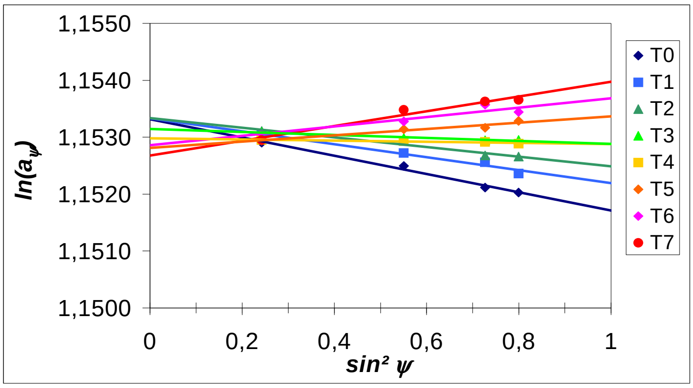

Figure 5 : $ln(a_\psi)$ versus $sin^2\psi$ plots concerning tungsten (310) peak of the W6/Cu18 multilayer for several increasing applied loads (T0 to T7).

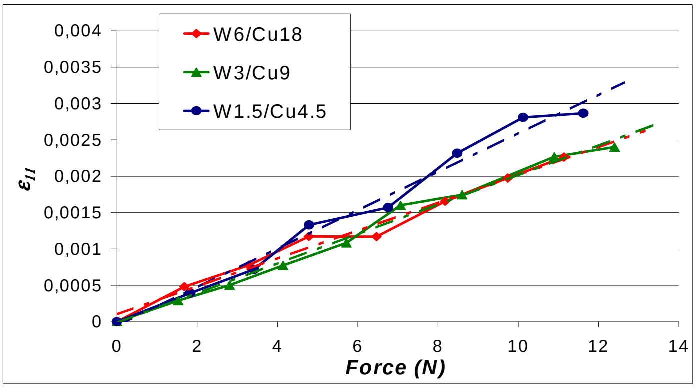

Figure 6: Applied strains ε_{11} extracted from the linear regressions on the $ln(a_\psi)$ versus $sin^2\psi$ plots (all ψ values for (310) peak), as a function of the applied load, for the 3 period values.

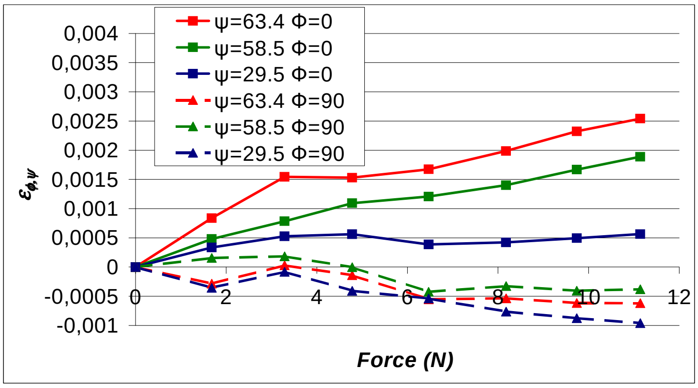

Figure 7: Evolution of the Cu(311) strain in the W6/Cu18 multilayer as a function of the applied load for 3 values of the ψ angle, along (*Φ*=90) and perpendicular (*Φ*=0) to the tensile axis .

Perpendicular strains have also been measured for W and Cu. Figure 7 presents a comparison between the longitudinal and transversal strains deduced from Cu(311) peak shifts. The observed evolutions are as expected. However, the strain in copper is larger than the strain in tungsten (Fig.4) whatever the force for ψ=63.4°. This difference may be due to local elastic anisotropy in copper.

Conclusions:

Preliminary data analysis shows promising results in terms of size effects on the mechanical behaviour of W/Cu multilayers. Indeed, figures 4 and 6 indicate a softening of tungsten layers in the case of the thinner periods. This softening needs to be confirmed and quantified by means of a thorough analysis of all experimental data (other diffracting plane families). It should be emphasised that strain measurements on the W1.5/Cu4.5 sample would not have been possible without a synchrotron X-ray source.

Concerning further experiments, more beam-time would be needed to study plastic yield thanks to diffraction peak broadening. Uncertainties may also be reduced by using improved jaws to guarantee that the specimen remains flat during its loading. Finally, the difficulties faced with copper (copper X-ray lines convoluted with tungsten ones) may lead us to change the “spacer” material, replacing Cu by another face-centred cubic metal like gold or silver; another idea consists in using anomalous scattering to reinforce the signal from one element or the other.

Let us mention that an oral presentation on W/Cu multilayer mechanical behaviour has been accepted at the MRS Fall Meeting to be held in Boston (USA), November 2006.