EUROPEAN SYNCHROTRON RADIATION FACILITY

INSTALLATION EUROPEENNE DE RAYONNEMENT SYNCHROTRON

ESRF

Experiment Report Form

The double page inside this form is to be filled in by all users or groups of users who have had access to beam time for measurements at the ESRF.

Once completed, the report should be submitted electronically to the User Office via the User Portal:

https://wwws.esrf.fr/misapps/SMISWebClient/protected/welcome.do

Reports supporting requests for additional beam time

Reports can be submitted independently of new proposals – it is necessary simply to indicate the number of the report(s) supporting a new proposal on the proposal form.

The Review Committees reserve the right to reject new proposals from groups who have not reported on the use of beam time allocated previously.

Reports on experiments relating to long term projects

Proposers awarded beam time for a long term project are required to submit an interim report at the end of each year, irrespective of the number of shifts of beam time they have used.

Published papers

All users must give proper credit to ESRF staff members and proper mention to ESRF facilities which were essential for the results described in any ensuing publication. Further, they are obliged to send to the Joint ESRF/ ILL library the complete reference and the abstract of all papers appearing in print, and resulting from the use of the ESRF.

Should you wish to make more general comments on the experiment, please note them on the User Evaluation Form, and send both the Report and the Evaluation Form to the User Office.

Deadlines for submission of Experimental Reports

Experimental reports must be submitted **within the period of 3 months** after the end of your experiment.

The deadlines for submitting a report to link to a new proposal ("relevant report") are:

- 1st March Proposal Round - **5th March**
- 10th September Proposal Round - **13th September**

Instructions for preparing your Report

- fill in a separate form for each project or series of measurements.
- type your report, in English.
- include the reference number of the proposal to which the report refers.
- make sure that the text, tables and figures fit into the space available.
- if your work is published or is in press, you may prefer to paste in the abstract, and add full reference details. If the abstract is in a language other than English, please include an English translation.

ESRF	**Experiment title:** *In situ* analysis of the structural evolution within thermally cycled Cu/W nano-multilayers	**Experiment number**: 28-01-1208
Beamline: BM28	**Date of experiment**: from: 06/04/2017 to: 09/04/2017	**Date of report**: 14/04/2017
Shifts: 9	**Local contact(s)**: Didier Wermeille	*Received at ESRF:*
Names and affiliations of applicants (* indicates experimentalists): **León Romano Brandt*** **Enrico Salvati*** **Chrysanthi Papadaki*** **Alexander M. Korsunsky**		

Report:

Diffusion in thin films can significantly differ from the bulk material behaviour [1]. Even though Copper and Tungsten are immiscible, diffusion processes can be observed at temperatures far below their melting point. The poor thermal stability of the layered structure can be referred to the proximity of a volume element to either a free surface or an interphase boundary [1]. It is therefore necessary to observe the material behaviour in situ to obtain a fundamental understanding of the kinetics of inter- and intra-layer diffusion processes as well as the structural evolution of the surface.

Copper/tungsten (Cu/W) nano-multilayer with the layer thickness of 18/6 nm, respectively, were deposited onto a Silicon substrate (for GISAXS and XRR) and a Kapton susbtrate (for WAXS) by ion beam deposition. The multilayer consists of ten periods of the copper/tungsten bilayer combination, with the total film thickness of 240 nm (**Fig. 1**). This combination of materials is particularly attractive for microelectronic applications [2], as tungsten provides an efficient diffusion barrier. The high thermal conductivity of Copper combined with the low thermal expansion coefficient of Tungsten allows thermal management on the microscale, for instance in heatsink applications [2]. The Cu/W combination is also used for radiation protection. It is therefore crucial to obtain a fundamental understanding of the thermal diffusion processes that occur when heating the multilayer.

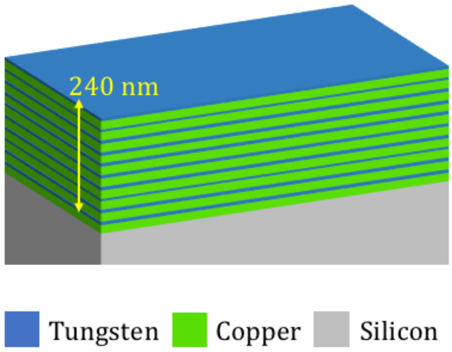

Fig. 1: Sample overview.

In order to advance this understanding, we proposed a multi-technique approach for an extensive characterisation of the above-mentioned diffusion processes. These techniques can be grouped in two categories. Firstly, our approach sensitive to the surface and interfaces uses the combination of Grazing-Incidence Small-Angle Scattering (GISAXS) and X-Ray Reflectivity (XRR). In addition, the phase composition and evolution within the material are analysed by Wide-Angle X-Ray Scattering (WAXS). For the temperature-dependent analysis of the properties, the sample was attached to the heating cell of a 400K furnace using Cerastil C7 ceramic paste. The furnace chamber was then pumped down to the vacuum of ~$2 \cdot 10^{-2}$ mbar.

The beamline setup of BM28 (XMaS) is extremely well suited for using the combination of these techniques. The Pilatus 300K 2D-detector was mounted at th e fixed distance of 1570mm from the sample for the acquisition of GISAXS frames. An APD KF16 point detector was mounted on the arm of the Huber diffractometer for XRR. For each acquisition of the 2D SAXS pattern the diffractometer arm was moved out of the sample-detector line, and GISAXS frames were acquired for different 2-theta angles. Afterwards, the diffractometer arm was moved back, and XRR patterns weer acquired in the scanning mode. This procedure was followed through different heating profiles. One continuous heating ramp was performed from 30˚C to 400˚C with frame acquisition for every 10˚C (**Fig. 2a**). Afterwards, a temperature cycle consisting of three heating and cooling periods 30˚C-250˚C-30˚C was performed with a larger frame step size (**Fig. 2b**).

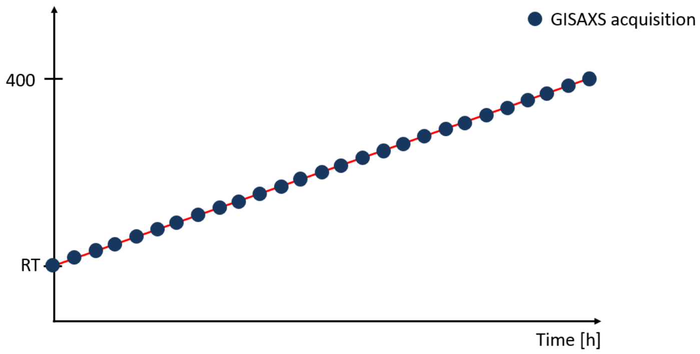

Fig. 2a: Constant heating ramp.

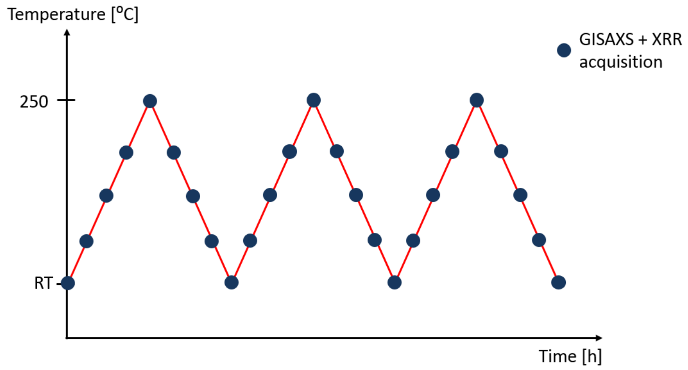

Fig 2b: Cycling temperature profile.

The GISAXS pattern evolution during the heat ramp is shown on **Fig. 3**:

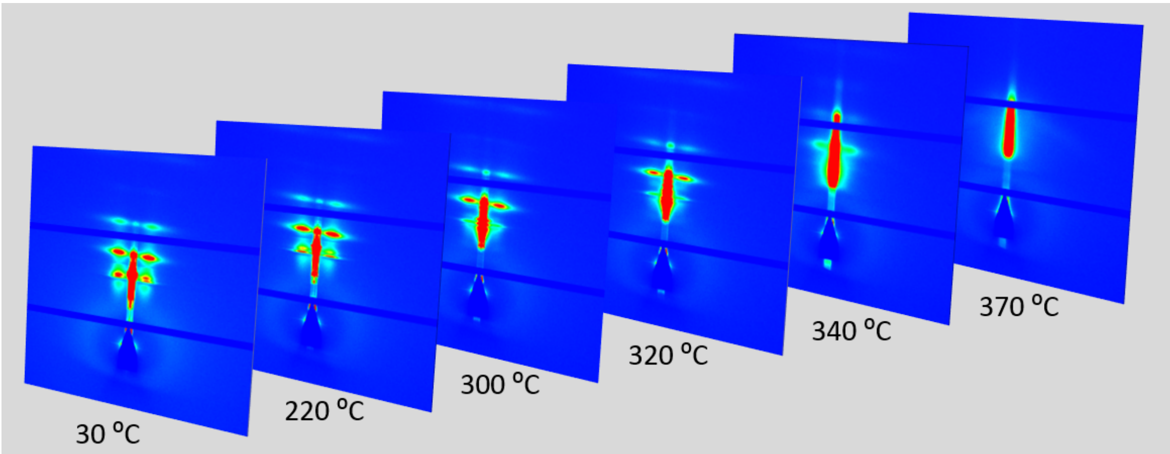

Fig. 3: GISAXS pattern evolution during constant heating ramp

The XRR profile showed the generation of new peaks and oscillations during thermal cycling, as can be seen in **Fig. 4**.

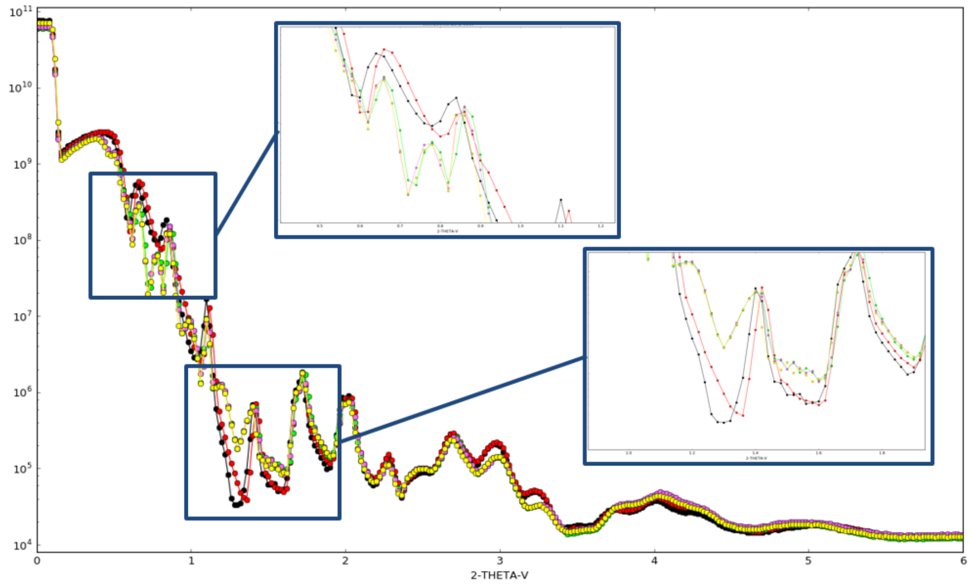

Fig. 4: XRR pattern evolution during thermal cycling: new oscillations appear at higher temperatures

For the WAXS setup, the sample was mounted on the heating cell with a thermally conductive copper frame (**Fig. 5a**) and the Pilatus 300K 2D-detector was mounted on the arm of the Huber diffractometer. The same temperature cycles were performed, while WAXS patterns were acquired for three different – partially overlapping - detector angles. An example of the composite pattern collected is shown in **Fig. 5b**. Changes in the WAXS pattern are observed during cycling, related to phase transformations within the multilayer.

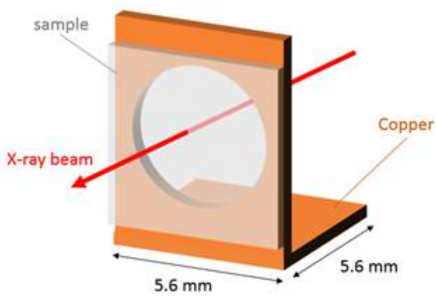

Fig. 5a: Sample mounting for transmission-WAXS on thermally conductive copper holder

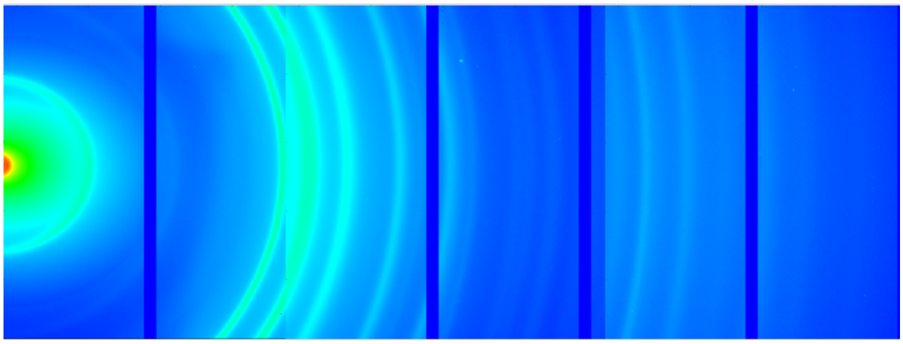

Fig. 5b: Example WAXS pattern obtained during in situ cycling

The combination of these technques will allow fundamental insights into thin film diffusion, including in immiscible materials, where an arrangement of nanoscale inclusions of distinct phase are formed. The detailed materials science implications of the observations will become clear once the full data sets are evaluated.

References

[1] Balluffi, R.W. and J.M. Blakely. "Special Aspects Of Diffusion In Thin Films". *Thin Solid Films* 25.2 (1975): 363-392. Web. 12 Sept. 2016.

[2] Girault, B. et *al*. "Cu Coverage Effect On W Crystallites Texture Development In W/Cu Nanocomposite Thin Films". *J. Appl. Phys.* 109.1 (2011): 014305.