EUROPEAN SYNCHROTRON RADIATION FACILITY

INSTALLATION EUROPEENNE DE RAYONNEMENT SYNCHROTRON

Experiment Report Form

	Experiment title: Time-resolved spatial local atomic fluctuations in metallic glasses studied by XRD-PDF-CT	**Experiment number:** HC 4089
Beamline: ID 11	**Date of experiment:** from: 18.02.2021 to: 22.02.2021	**Date of report:** 2.08.2021
Shifts: 12	**Local contact(s):** Carlotta GIACOBBE	*Received at ESRF:*

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Report:

The experiment was performed after a long break that included the upgrade shut-down of the ESRF and the Covid-19 pandemic.

The actual experiment is a follow-up of an older experiment (HC3830), which had as a main output the publication titled "**X-Ray Diffraction Computed Nanotomography Applied to Solve the Structure of Hierarchically Phase-Separated Metallic Glass**", *ACS Nano* 2021, 15, 2386−2398, and which was considered as a Spotlight in Science by ESRF: https://www.esrf.fr/home/news/spotlight/content-news/spotlight/spotlight383.html.

In regard with the previous experiment, this new one took in account the power of the new EBS source and it was designed for that. If the previous experiment was a **static** one, now we moved toward a **dynamic**, time-resolved experiment. As a simple detail, in the previous experiment one complete nano-CT scan took 10 hours, while with the new EBS this time was reduced to 45 minutes. In the following the detailed description of the scientific case and the output is presented.

The experiment aimed to resolve the local atomic fluctuation in metallic glasses (MGs). The (final) key focus is the influence of atomic contributions on the macroscopic properties such as plasticity or extremely soft magnetic behavior, hence detailed analysis of the corresponding structural variations in the medium and short range order is necessary. The **key point** is to detect the **sub-micron local fluctuations** and determine their nature and evolution upon heating, because this is the crucial aspect in order to **understand** and to **control** the behavior of MGs. X-ray diffraction (XRD) data, taken in computer tomography (CT) configuration, allow the analysis of the structure at the nanoscale. In order to increase the resolution, the beam was focused down to 500 nm and the sub-micron resolution was attained via the number of scans. As a standard sample we used a hierarchically phase-separated $Gd_{27.5}Hf_{27.5}Co_{25}Al_{20}$ metallic glass (MG). Then we investigated $Pd_{77.5}Cu_{6}Si_{16.5}$, $Fe_{68.352}B_{23.04}Y_{4.608}Nb_{4}$ and $Fe_{69.54}B_{20.9}Y_{4.56}Mo_{5}$ bulk metallic glass (BMGs) samples.

The current experiment is a continuation of the successful HC3830 experiment, in which we demonstrated the reliability of XRD-PDF computer tomography with nanometer resolution (XRD-PDF-nCT). The new synchrotron setup, in which the X-ray flux is order of magnitude higher than before, and the new zero readout time detector from ID 11 facilitated the **dynamic experiment**, i.e. the temperature evolution of the structure. This contributes not only to the **basic understanding** of MGs behavior but will allow as well to define **new annealing strategies**. The corresponding heating curves for MG samples are presented in Fig. 1. Basically, the MG sample was heated up to the desired temperature (under vacuum, by using a homemade- ESRF- device) and then cooled down to room temperature. Further, a complete nCT scan was performed. This MG sample was considered as a standard sample, in order to prove the reliability of the method. Further, the BMG samples were **continuously** heated and the nCT scan performed at high temperature (so a real **dynamic** experiment). The chosen temperature for the phase-separated MG were chosen in such a way that, at a certain moment, the sample became composite, i.e. instead of two hierarchically amorphous phases one crystallized. This was done in order to assess the best resolution that can be obtained by using this method.

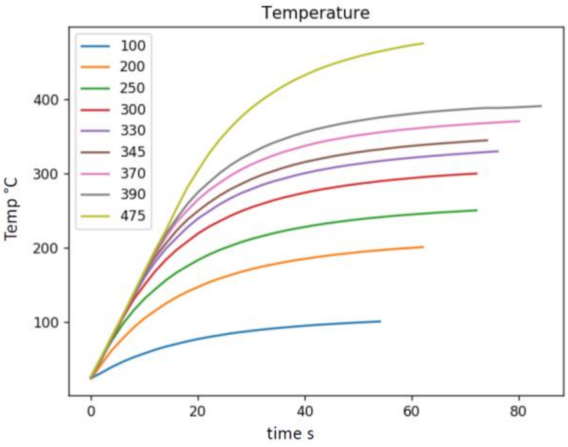

Fig. 1 Heating curves for MG sample

Fig. 2 shows the reconstructed cross-section of the phase-separated MG at room temperature. It was demonstrated this alloy shows upon separation two amorphous phases, one (Co,Hf)-rich and one Gd-rich. There are clear differences and, as it can be seen, the Gd-rich phase seems to form the matrix. This is perfectly in-line with the previous findings. Fig 3 shows for comparison the same sample, this time measured at 475°C.

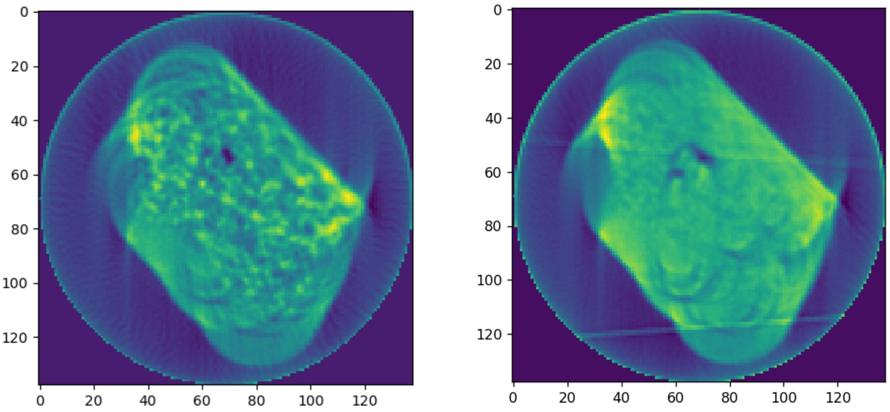

Fig. 2 The reconstructed cross-section (room temperature) of the MG sample, details upon (Co,Hf)-phase (left) and Gd-phase (right).

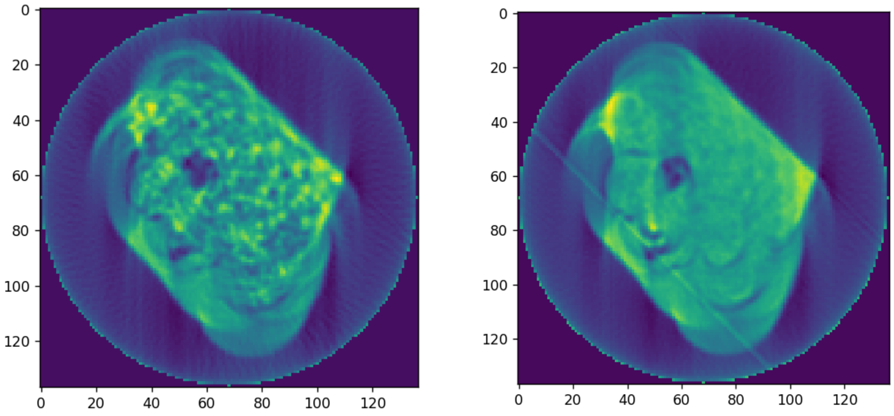

Fig. 3 The reconstructed cross-section (at 475°C) of the MG sample, details upon (Co,Hf)-phase (left) and Gd-phase (right).

At this temperature the (Co,Hf)-rich phase is in the nanocrystalline state, which is clearly seen in the picture. The cross-section seems to be coarser than the corresponding one at room temperature.

The data corresponding to the BMGs are under analysis and two scientific publications are currently in preparation.