ESRF	**Experiment title:** Temperature dependence study of high-frequency dynamics of fragile metallic glasses by IXS	**Experiment number:** HD-248
Beamline: ID28	**Date of experiment:** from: May 14th, 2008 to: May 20th, 2008	**Date of report:** September 20th, 2008
Shifts: 18	**Local contact(s):** Mathieu Le Tacon	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):
Daniel Crespo, Pere Bruna, Eloi Pineda, Jorge Serrano, Trinitat Pradell (*,1)
(1) Universitat Politecnica de Catalunya, Campus del Baix Llobregat, Castelldefels, Spain

Report:

Metallic glasses represent a new class of amorphous alloys relevant in the field of structural applications, as alternative to standard steels, and for applications in corrosion resistant coatings, micromechanics, and biomedicine, among others.

The goal of the experiment HD-248 was to ascertain the change in fragility upon change in temperature in two samples of Pd-based alloys synthesized using different cooling rates. To that aim, inelastic x-ray scattering experiments were performed at the beamline ID28, in order to extract information about the sound speed and the non-ergodicity factor, the latter accesible via the ratio between elastic and inelastic scattering signal [1].

We employed 18 shifts of 16 bunch mode synchrotron radiation in order to perform such study, using monochromatic x-rays of 17.794 keV. Despite technical difficulties due to a decrease in ring current of about 30%, we succeeded to obtain good quality IXS data for $Pd_{77}Si_{16.5}Cu_{6.5}$ melt spun ribbons at both room temperature and 200K, using a closed-cycle ST15 cryostat. This composition shows a large fragility index, $m = 75$, as determined from viscosity measurements, which is very close to the value of $m_{IXS} = 63(9)$ that we obtained using the relation between non ergodicity factor and fragility reported by Scopigno and coworkers [1]. Furthermore, we performed a test measurement on a strong Ce-based metallic glass, $Ce_{70}Al_{10}Ni_{10}Cu_{20}$, which turned out to show much stronger IXS signal than expected, thus yielding directly publishable data. This alloy has a fragility index $m = 23$, as determined by calorimetric measurements, and we obtained similar values from the non-ergodicity factor ($m_{IXS} = 21(9)$).

The longitudinal sound speed determined from the IXS experiments for both compositions is 10% larger than the values reported in the literature, stemming from ultrasound experiments. This result is in agreement with a similar behavior reported for $Pd_{42.5}Ni_{7.5}Cu_{30}P_{20}$, where an ansatz of two mechanically different nanoscale domains coexisting in the metallic glass was proposed [2]. In order to shed more light on the validity of this hypothesis, higher resolution IXS experiments should be conducted using the 11 11 11 reflection in the monochromator, and a higher photon flux is required. We therefore posponed this issue for a continuation proposal.
We can summarize the main results of the IXS experiment in the following items:

- We have determined the dispersion relations of longitudinal acoustic modes for $Pd_{77}Si_{16.5}Cu_{6.5}$ and $Ce_{70}Al_{10}Ni_{10}Cu_{20}$ metallic glasses. They represent benchmark systems for fragile and strong glasses, respectively.
- The derived sound speeds exceed by about 10% the values reported from ultrasound experiments. The origin of this different behavior remains to be elucidated, and is accessible via higher resolution IXS experiments.
- The spectral linewidths show a linear dependence with peak energy, in the 2 nm^{-1}–Qmax/2 momentum transfer region, where Qmax is the momentum transfer corresponding to the peak in the static structure factor.
- The excellent quality of the collected spectra allows us to determine the nonergodicity factor and verify its relation with the fragility of the glass forming liquid. This relation is shown to hold for both strong and fragile metallic glasses.
- IXS data obtained for $Pd_{77}Si_{16.5}Cu_{6.5}$ at 300K and 200K result in similar IXS-derived fragility values, therefore pointing out negligible anharmonic contributions up to room temperature.

The results obtained in this experiment have been reported in the 15^{th} International Symposium on Metastable, Amorphous and Nanostructured Materials (ISMANAM-2008) and will be partially published in J. Alloys and Compounds. Another manuscript is in preparation.
References:
[1] T. Scopigno, G. Ruocco, F. Sette, and G. Monaco, Science 203 (2003) 849.
[2] T. Ichitsubo *et al.*, Phys. Rev. B 76 (2007) 140201.
[3] T. Scopigno *et al.*, Phys. Rev. Lett. 96 (2006) 135501.