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| | Experiment title: Atomic dynamics in beta-relaxation-dominated metallic glasses studied by x-ray photon correlation spectroscopy | Experiment number: HC1426 |
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| Shifts: | Local contact(s): B. Ruta and Y. Chushkin | <i>Received at ESRF:</i> |
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Report:

By using X-ray Photon Correlation Spectroscopy (XPCS) at the ID10 beamline, ESRF, we have measured the atomic dynamics in La-Al-M (M=Ni and Cu) metallic glasses (MGs) under different temperatures in the glassy state ($T_g = 395$ K for LaAlCu and 429 K for LaAlNi MG). The data were collected for a unique wave vector q_0 corresponding to the position of the first sharp diffraction peak in the static structure factor, where q_0 is of about $2.11\text{-}2.12 \text{ \AA}^{-1}$ for these MGs. In this way we have been able to look directly at the atomic dynamics in metallic glasses, which is still difficult to access by using any other techniques.

The data have been collected in a wide angle configuration by two two-dimensional detectors placed vertically symmetric with respect to the incoming beam. Two detectors recorded simultaneously the signal scattered at the same wave vector, therefore leading to a better statistics and fast measurements. Sets up to ~ 1300 images have been collected with an exposure time of 3 s per image, depending on the investigated temperature.

A picture of the dynamics can be entirely captured by XPCS through the determination of the two-time correlation function $G(t_1, t_2)$ (TTCF) which represents the instantaneous correlation of the intensity I at two times t_1 and t_2 , being:

$$G(t_1, t_2) = \frac{\langle I(t_1)I(t_2) \rangle_p}{\langle I(t_1) \rangle_p \langle I(t_2) \rangle_p}$$

Here the average is done over all the pixels of the entire CCD. Figure 1 (left) shows a TTCF measured in the glassy state for $T = 333$ K. This quantity represents the instantaneous correlation between two times. The main diagonal from the left bottom corner to the up right corner indicates the experimental time, while the

intensity perpendicular to the main diagonal is proportional to the structural relaxation time. The line width along the main diagonal remains constant with time and it is relatively thin. This means that the atomic dynamics in the glassy state is governed by a fast, stationary, structural relaxation when the temperature is far below the T_g with a characteristic time of few hundreds of seconds.

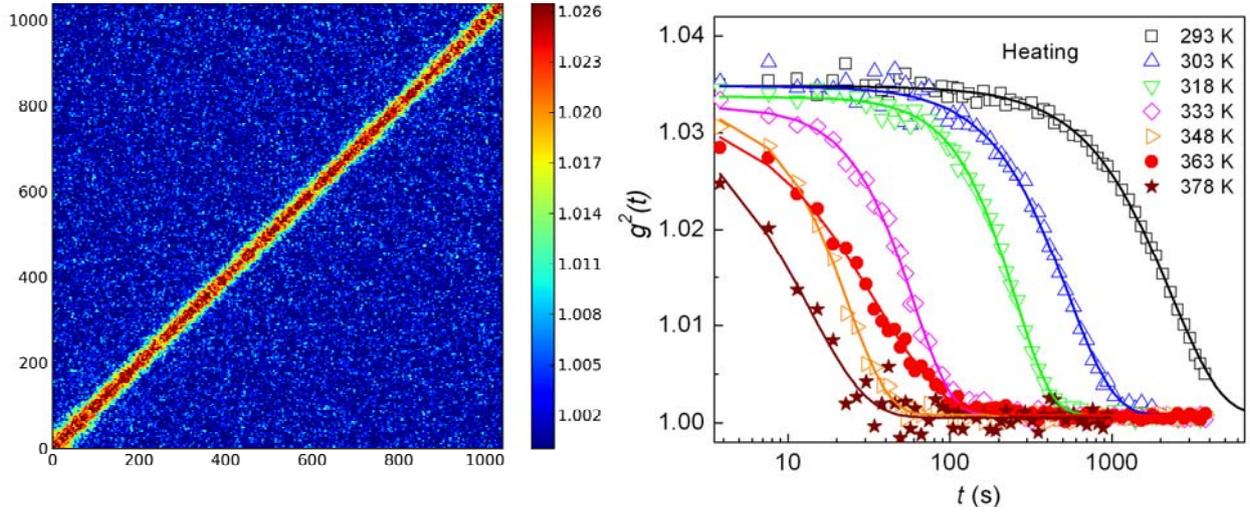


Figure 1 (Left) Two times correlation function measured with XPCS for the ascast LaAlNi glass at 333 K, far below the glass transition temperature (429 K). The axis corresponds to the number of collected images with an exposure time of 3 s for each. (Right) Temperature dependence of correlation functions measured at $q_0 = 2.11 \text{ \AA}^{-1}$ for the ascast LaAlNi MG. The lines are the best fits using the KWW model function.

Another alternative representation is the autocorrelation function,
$$g_2(q, t) = \frac{\langle\langle I_p(q, t_1) I_p(q, t_1 + t) \rangle_p \rangle}{\langle\langle I(q, t_1) \rangle_p \langle I(q, t_1) \rangle_p \rangle},$$

where $\langle\langle \dots \rangle_p$ denotes the ensemble average over all the pixels of the detector and $\langle \dots \rangle$ is the temporal average. Fig. 1(right) shows the temperature dependence of the correlation functions in the as cast LaAlNi MG on heating from the glassy state. Upon increasing the temperature the dynamics shifts towards faster time scales due to the thermal motion but at 363 K where an anomalous slowing down of the dynamics is observed.

Additional measurements were performed in both La-Al-M (M=Ni and Cu) MGs following different thermal protocols on both as cast and fully pre-annealed samples and the analysis is ongoing. Combining the XPCS data with DSC, DMA and also in situ XRD and XAFS results, we hope to clarify the detailed structural and dynamical changes occurring in both MGs with temperatures.