	Experiment title: Probing dynamical heterogeneities in glasses by small-angle nuclear resonant scattering	**Experiment number**: SC-5288
Beamline: ID18	**Date of experiment**: from: 13/07/2022 to: 19/07/2022	**Date of report**: 12/07/2022
Shifts: 18	**Local contact(s)**: Dimitrios Bessas, Aleksandr Chumakov	*Received at ESRF:*

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Report:

Driven by scientific reasons, the experiment SC-5219 (Main proposer Caporaletti), was exchanged with the experiment SC-5288 (Main proposer Martinelli) accepted for 2022/I, in agreeement with the User Office and the ESRF Safety. Both proposals had several proposers in common.
For this reason, the measurements envisaged for experiment SC-5219 were performed during this beamtime and the following report describe the corresponding experimental results. Conversly, the report of experiment SC-5219 show to the results of proposal SC-5288.

The aim of the experiment was to investigate the dynamic hetereogenities (DH) in glasses across their glass-transition temperature by employing small-angle nuclear resonant scattering (NRSAXS). Dynamic hetereogenities consists of microscopic spatio-temporal fluctuations in the dynamics supercooled liquids, which are expected to be correlated over a distance of roughly 1 nm, close to the glass-transition. DH and, in particular, the associated correlation lengths, are expected to play the role of order-parameter in the ideal glass-transition [1], though their observation is usually elusive to most experimental techniques.
We wanted to measure the DHs, by probing the associated hetereogenities in the atomic mean square displacement $\langle x^2 \rangle$. The cross section for nuclear resonant elastic scattering depends in fact on the Lamb-Mossbauer factor (f_{LM}), which, differently from the Debye-Waller factor (f_{DW}), does not depend on the exchanged wave-vector but only on the incident one [2]. So, in the forward direction (small angle scattering range), it will not be close to unity as usually observed in conventional X-ray scattering and

consequently it should be sensitive to spatial fluctuations of $\langle x^2 \rangle$ the same way X-rays small-angle scattering does with density.

The sample initially selected for the measurements was the metallic glass-former was (57Fe0.66Nb0.04B0.3). However this sample displayed a very fast crystallization kinetics above its glass-transition temperature, which made difficult collecting statistics at high temperatures. For this reason, we measured, as backup, the silicate glass-former $Na_2{}^{57}FeSi_3O_8$ (95% enriched, T_g=744K) [3]. The measurement was performed by placing the APD detectors at 1 m from the sample and by scanning vertically the beam-profile. Slits were placed before and after the sample to achieve a q-resolution of roughly 0.05nm^{-1} and we scanned a q-range from 0 up to 5 nm^{-1}. Experimental and theoretical works suggest DH to have a characteristic size of roughly $\epsilon \simeq$2-5nm [1], so we expected to observe a contribution to the small-angle scattering at q-values of roughly $\frac{1}{\epsilon} \simeq 0.2 - 0.5$ nm^{-1}. However, the angular profile does not display any major feature in such range. On the contrary we found a tail at much lower q-values (0.02-0.06 nm^{-1}), which increased upon heating above the glass-transition temperature. Such smaller q-values suggest the presence of hetereogenities with a much longer scale than DH, which could be of either structural of dyanamical origin. We ruled out a contribution from crystallization by monitoring the static structure factor of the glass before and after each measurements. Future experiments are clearly required to determine the origin of the observed anomalous NRSAXS. In particular it would be crucial to increase the angular resolution in the region of interest and reduce the contribution of air scattering. Both improvements can be easily achieved by using, in detection, a crystal reflection as analyzer, which should allow us to probe more effectively this smaller q-range [2].

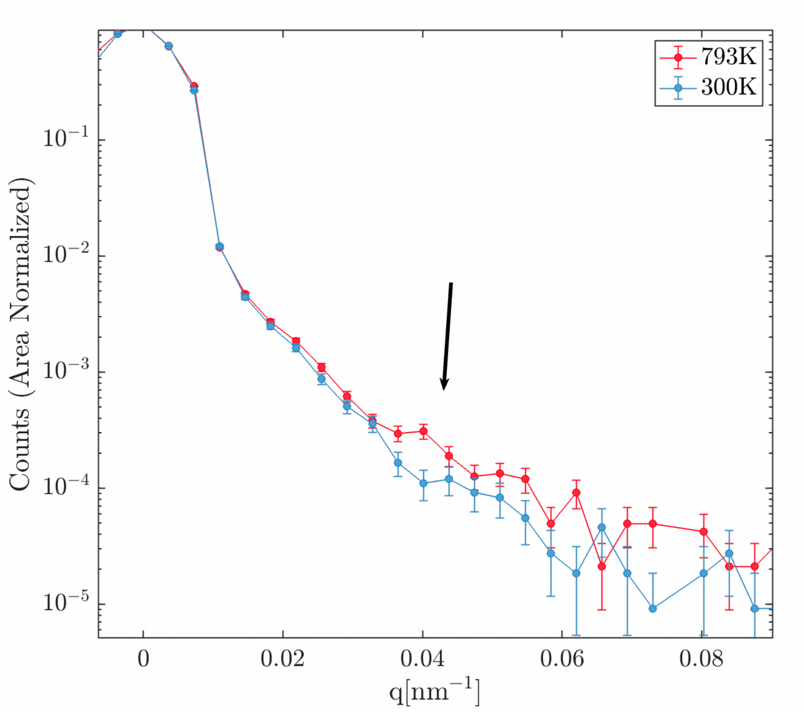

Figure 1Figure 1: Angular dispersion of nuclear resonant scattering as measured from Na2FeSi3O8 at 300K (blue circles with errorbar) and at 793K (red circles with errorbar). A hint of enhanced resonant scattering intensity at 793K can be seen in the range from 0.02nm^{-1} up to 0.06 nm^{-1}

References

[1] Berthier, L.; Biroli, G. Rev. Mod. Phys 83, 587− 645 (2011)
[2] Yu. V. Shvyd'ko et al., Phys. Rev. B, 54, (1996).
[3] A. Monaco, Phys. Rev. Lett. 97, 135501 (2006).