ESRF	**Experiment title:** Probing the structural relaxation of an oxide glass	**Experiment number**: HC3748
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Report:

During the experiment HC-3748, "Probing the structural relaxation of an oxide glass", we used x-ray photon correlation spectroscopy (XPCS) to investigate the structural relaxation above the glass transition temperature in the prototypical glass former B_2O_3. It is well understood that in oxide glasses [1, 2], and also in boron-oxide [3], a peculiar effect is observed with XPCS deep in the glassy state: the intermediate scattering function (or the density correlation function) decorrelate with a typical relaxation time (τ) in the order of tens to hundreds of seconds, despite the structural relaxation is expected to be frozen in. This effect appears to be flux dependent, with $\tau \sim 1/F$: the incident x-rays triggers density rearrangements and, at the same time, they probe this relaxation. In this experiment, we aimed at overcoming this beam-induced phenomenon and probe the intrinsic structural relaxation of a glass former, investigating the exchanged wave-vector dependence, q, across the peak of the structure factor at a temperature where the structural relaxion is faster than the beam induced process.

The experiment was conducted on two different kind of samples: pure vitreous B_2O_3 and a lithium borate glass, $(Li_2O)_{0.5}(B_2O_3)_{0.5}$. The initial plan of the experiment was to measure the wavevector dependence of the dynamics in the pure boron oxide glass. However, due to the fast beam induced motion in this sample, we decided to probe the q dependence in the lithium borate glass, where the decorrelation time induced by the beam is almost an order of magnitude slower.

XPCS investigation of the pure boron oxide glass

The samples were 100 μm thick B_2O_3 disks, placed in an electric furnace with Kapton windows. The incident energy was set to 8.1 keV, implying the peak of the structure factor near $\theta \sim 20$ degrees ($q = \frac{4\pi}{\lambda} \sin\frac{\theta}{2}$). The chosen detector for the experiment was the EIGER 500K, a new generation of x-ray detectors allowing high

frame rate (22 kHz in 4-bit mode) with pixel size of 75 um. The detector allowed us to probe typical relaxation times of less than one second, with exposures down to 20 ms. The sample's temperature was increased up to the liquid phase, checking the scaling of the relaxation time with the flux and the behavior as a function of temperature. Indeed, previous studies shown that a clear relationship exists between the induced and the structural relaxation time as a function of temperature [3].

An example of the typical autocorrelation function obtained for the B_2O_3 glass at 310°C is reported in figure 1. As can be clearly seen, a good data quality is achieved with an exposure time of 50ms and averaging 18 scans of 10000 images each one. The obtained relaxation time in this particular case is $\tau = (3.43 \pm 0.15)\ s$.

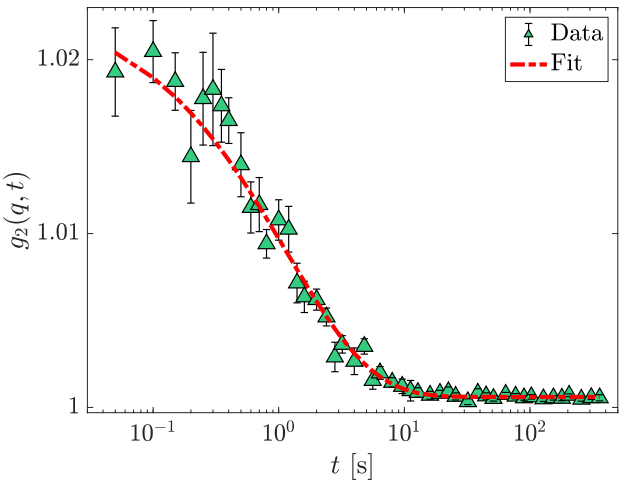

Fig.1: Example of correlation function obtained for the B_2O_3 glass on the peak of the structure factor, q~15 nm^{-1}, at the temperature 310°C. The exposure time for this particular measurement was set to 50ms and the total number of images is $1.8 \cdot 10^5$. Figure from ref. [4].

However, both because of the low scattered intensity and the need to measure a reasonable part of the decay in fast autocorrelation functions (thus reducing the exposure time), we needed to increase the number of images per point up to ~$5 \cdot 10^5$.
The results of the first part of the experiment are under publication [4].

XPCS investigation of the lithium borate glass

Because of the difficulty to obtain a good signal to noise ratio for the pure boron oxide in the proximity of the region were the spontaneous sample dynamics is visible, mainly because of the fast beam induced dynamics, we decided to exploit the last two days of beamtime to probe the XPCS signal of a lithium borate, $(Li_2O)_{0.5}(B_2O_3)_{0.5}$, a glass where the beam-induced dynamics is slower by a factor ~10.

Data analysis is still ongoing in order to extract information about the *q*-dependence of the relaxation time. Preliminary results show that in the beam induced dynamics region (below the glass transition temperature), the relaxation time scale as a power law of the scattering vector *q*, in agreement with what was found for silica glass [2]. In the undercooled liquid phase, where the contribution of the induced dynamics is negligible [3], this divergence seems to disappear with the dynamics becoming progressibvely q independent, marking a transition between spontaneous and induced density rearrangements.

This is one of the very first experimental investigations of the wavevector dependence of the structural relaxation time in an oxide glass close to the glass transition temperauture. We believe that further measurements are necessary to properly establish this important result in a wider q range and with higher accuracy.

[1] B. Ruta *et al.*, Nat. Comm, **5**, 3939 (2014).
[2] B. Ruta *et al.*, Scientific Reports **7**, 3962 (2017).
[3] G. Pintori *et al.*, Phys. Rev. B **99**, 224206 (2019).
[4] A. Martinelli *et al.*, Phil. Mag. (2020 accepted for publication).