ESRF	**Experiment title:** Topology driven elastic transitions in SiO_2-$CaAl_2O_4$ glasses	**Experiment number**: HC3445
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Report:

Following the original proposal, a series of $xCaO - xAl_2O_3 - (100-2x)SiO_2$ glass plates of a few mm and ~20-40 μm in thickness were prepared as a compromise between scattering intensity and contrast at the fixed incident energy of 8 keV. We further prepared a second series of $20CaO - yAl_2O_3 - (80-y)SiO_2$ similar plates. The main aim of the proposed experiment was to evidence a composition-driven change of beam-induced dynamics when crossing an expected rigidity transition around the $CaAl_2Si_3O_{10}$ glass composition (x=y=20).

Within the allowed beam time, eight of these CAS glasses have been studied at room temperature. The plates were put in a vacuum evacuated sample container placed into the beam. We first measured the static structure factor S(Q) and time-series of speckle patterns at the first maximum Q_p of S(Q) using the standard wide-angle setup. Speckle patterns were detected with an IkonM CCD from Andor Technology (1024×1024 pixels, 13×13 μm^2 pixel size) installed perpendicularly to the horizontal scattering plane, ~70 cm downstream of the sample. In this way, the detector covers a solid angle corresponding to a Q resolution of about 0.04 $Å^{-1}$.

A typical example of the obtained two-time correlation functions is shown in figure 1 for the x=5 glass. The contrast is very good for a silicate glass, about 6% at Q_p = 1.53 $Å^{-1}$. A slight broadening of the diagonal can be detected with time indicating an evolution of the measured dynamics. The latter is better seen in Figure 2 where the normalized intensity auto-correlation function $g_2(Q,t)$ at two different irradiation times (yellow squares in Figure 1) is calculated with a constant time-window of 900 s, corresponding to 250 frames. On a fresh spot (green open triangles), the decay is well described by a compressed exponential, $g_2(Q,t) = 1 + ce^{-2(t/\tau)^{\beta}}$, where the shape exponent parameter $\beta \approx 1.7$ and $\tau \approx 80$ s. At longer times, the shape differs as β decreases to 1.2 and τ increases to about 140 s. It clearly evidences that in these ternary glasses the beam-induced dynamics is not stationary but evolves with irradiation time. We also verified that this behavior is similar at all Q values between 0.6 and 2.0 $Å^{-1}$.

Interestingly we found that the beam-induced dynamics evolves with the glass composition indicating a change around the expected x=y=20 composition. The analysis of the large amount of data is still ongoing but we are confident that these results will strongly improve our knowledge on the glassy state and will surely deserve to be published.

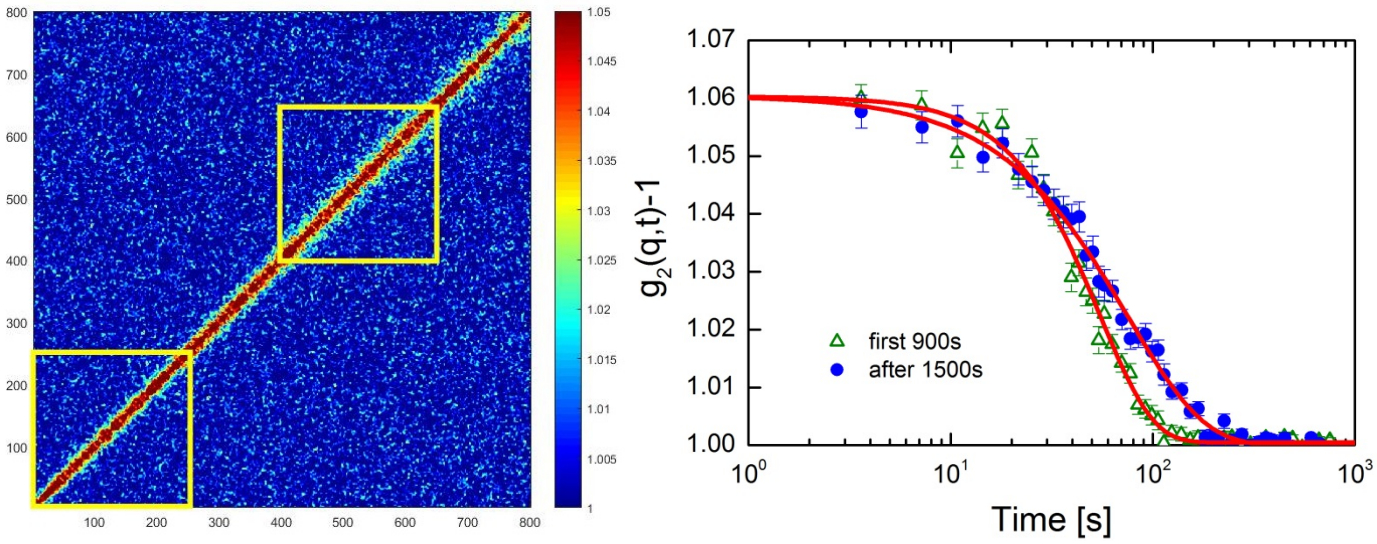

Figure 1: (left) Two-time correlation function measured at room temperature at the maximum structure factor S(Q) on the x=5 CAS sample. Left to right in frame number (Δt = 3.6 s). The yellow squares indicate the frames used to calculate the correlation curves plotted in Figure 2.

Figure 2: Normalized intensity auto-correlation functions at two selected irradiation times from Figure 1. Red lines are fits with a Kohlrausch-Williams-Watts function.

Finally, we used the last day to test the feasibility of the study of the intrinsic structural relaxation time at temperature close to T_g. To that effect, we chose the x=35 composition which is characterized by the slowest beam-induced dynamics of the CAS series ($\tau \approx 3000$ s). Approaching T_g from below we have indeed detected a sudden decrease of the measured relaxation time. This indicates that the intrinsic structural relaxation time takes over the beam-induced dynamics. For that glass, it clearly gives the unique opportunity to study the structural dynamics at the atomic scale over more than three decades in time across the glass transition region using the Eiger detector.