ESRF	**Experiment title:** **Elucidating the yielding of glasses by combining SAXS and rheology**	**Experiment number**: 26-02 715
Beamline: BM26B	**Date of experiment:** from: 23/01/2015 to: 26/01/2015	**Date of report:** 02/07/2015
Shifts: 9	**Local contact(s)**: Giuseppe Portale	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):

D. V. Denisov[1*]. M. T. Dang[1], B. Struth[2], P. Schall[1]

[1]Instiute of Physics, University of Amsterdam, Netherlands
[2]Photon Science Detector Systems, DESY, Germany

Report: The yielding of glasses is important for a wide range of materials including metallic glasses, polymer- and soft glasses. Yielding demarcates the property of any solid to flow and deform irreversibly under applied deformation [1]. At small applied stress and strain, the material deforms mostly elastically; at larger strain, the material starts to flow irreversibly, resulting in permanent deformation. We use a novel experimental setup with vertical x-ray beam which combines x-ray scattering technique and rheology to perform the oscillatory shear of a colloidal glass and monitor the structure factor evolution during the increasing strain amplitude [2].

The installation of new experimental setup with vertical x-ray beam took longer time than we expected and 1.5 days of measurements were spent for the calibration and beam alignment. Due to the reflection procedure to redirect the beam in the vertical position the intensity of the scattered beam was much lower than expected. Still part of the intended research was successfully carried out, where we have probed the samples response at different oscillation frequencies.

The experimental sample is a suspension consisting of silica particles with a diameter of 50 nm and a polydispersity of 10%, suspended in water. A small amount (1mM) of NaCl is added to screen the particle charges. The samples were prepared close to the glassy state with volume fractions 54-62%. We have used the rheometer to apply oscillatory strain with frequencies f=0.5-5 Hz and strain amplitude γ_0 increasing from $\gamma_{0min}=10^{-4}$ to $\gamma_{0max}=1$. We measure rheological macroscopic parameters (viscosity, storage and loss moduli) to track the sample transition from elastic state (low amplitudes) to plastic deformation (high amplitudes). We simultaneously monitor the scattered intensity at a frame rate of 1 Hz. From the recorded intensity, we determine the structure factor S(q) and focus on the first peak of the structure factor to elucidate changes in the nearest-neighbor structure upon yielding (Figure 1, left). By tracking the distribution of intensity along first scattering ring with respect to angle α we detect a weak two-fold symmetry imposed the applied shear (Figure 1, right). However this anisotropic symmetry only exist at low strain amplitudes; for amplitudes higher than a certain threshold γ_0^* the anisotropy vanishes signifying the transition of the sample into dynamically melted isotropic liquid state.

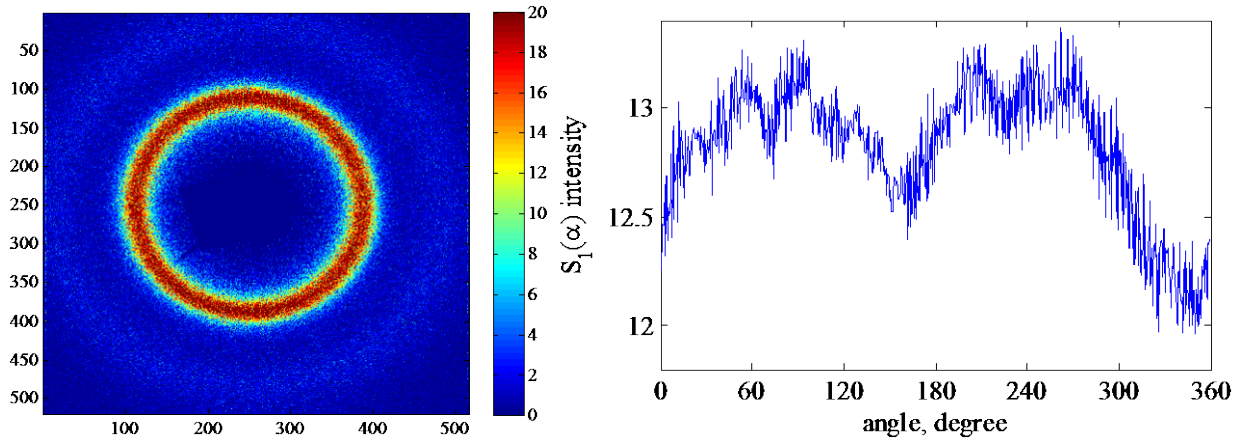
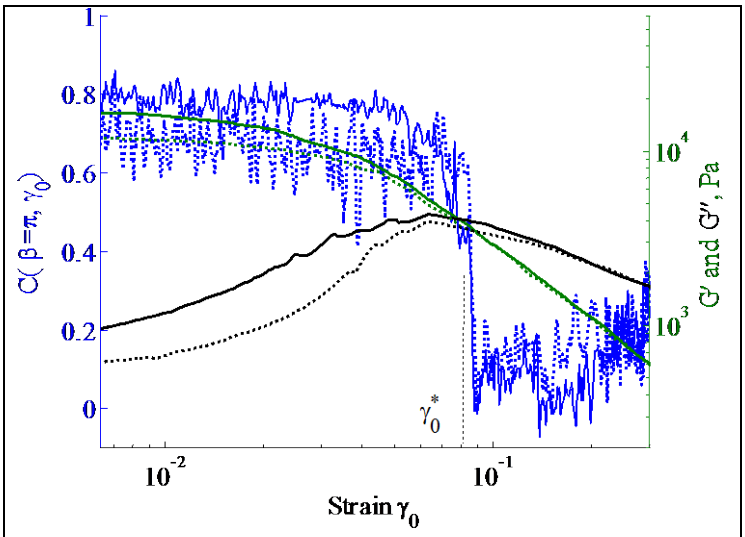


Figure 1: (Left) Scatter pattern of Silica colloidal glass. (Right) Evolution of structure factor S_1 along the angle α. With applied oscillatory strain the two-fold symmetry is detected.

By using specific techniques we can correlate the intensity along the angle α to remove the influence of the fluctuation ($C(\beta)=\langle S_1(\alpha+\beta)S_1(\alpha)\rangle_\alpha$). Alternatively we can study the fluctuation themselves by calculating the Kurtosis along the ring. Amazingly both quantities show sharp changes at amplitude γ_0^*, corresponding the glass transition from solid to liquid state (Figure 2).



Left axis: $C(\gamma_0, \beta=\pi)$ as a function of increased strain amplitude γ_0 (solid blue curve) and decreased strain amplitude γ_0 (dashed blue curve). Right axis: elastic and viscous moduli, G' and G'' (green and black).

During the experiments we have concentrated on the transition point γ_0^* behavior with respect to the driving frequency and volume fraction. We have found that γ_0^* weakly decreases with volume fraction in glassy state (φ>58%). From the other hand in the tested range of driving frequencies 0.5-5Hz the transition point slowly increases from ~10% to a maximum ~15%.

More interestingly we have confirmed that at transition point γ_0^* microscopic structure parameters always have a sharp change which coincides with the crossing point of G' and G'' (storage and loss moduli), which corresponds to sample transition from liquid the solid state on macroscopic level. Our measurements at ESRF have confirmed that this transition is reversible – by changing the direction of the oscillatory amplitude ramping from increasing to decreasing we can effectively start with dynamically liquidized glassy sample and turn it back to the solid state at γ_0^*. The transition point from solid to liquid and liquid to solid shows weak hysteresis behavior which we are going to explore in future works.

References

[1] H. A. Barnes, J. Non-Newtonian Fluid Mech. **81**, 133 (1999).

[2] D. Denisov, M. T. Dang, B. Struth, G. H. Wegdam, and P. Schall, Sci. Rep. **3**, 1631 (2013).