Local structure and dynamics of colloidal glasses probed with scanning small angle x-ray scattering

Liu, A.[1], Bojesen, E.D.[2], de Jonge, M.D.[3], Tabor, R.F.[4], Petersen, T.C.[5], Mudie, S.T.[3], Zaccone, A.[6] and Harrowell, P.[7]

[1] Monash Centre for Electron Microscopy and School of Physics and Astronomy, Monash University, Clayton, Australia, [2] Monash Centre for Electron Microscopy, Monash University, Clayton, Australia, [3] The Australian Synchrotron, ANSTO, Clayton, Australia, [4] School of Chemistry, Monash University, Clayton, Australia, [5] School of Physics and Astronomy, Monash University, Clayton, Australia, [6] Department of Chemical Engineering and Biotechnology, University of Cambridge, United Kingdom, [7] School of Chemistry, University of Sydney, Sydney, Australia

The structure of glasses is notoriously hard to measure, but local structure and symmetry in disordered materials is still expected to be key to understanding their nature and behaviours [1]. Recent theories have hypothesized that non-centrosymmetric local structures in glasses have larger amplitude dynamics and undergo non-affine transformations under shear due to unbalanced forces [2]. Shear transformations in these soft spots can avalanche, causing the well-known brittle failure of glasses.

We have shown that new order parameters accessible via small probe scanning transmission diffraction measurements can be used to measure the symmetry of nearest-neighbour clusters in glasses [3-6]. In this submission we extend our analysis to mapping local dynamics and local structural change under the application of a shearing force. In part A of the figure we show a small angle x-ray scattering (SAXS) pattern from a 300 nm colloidal glass with the beam size limited by a 7 μm near-field aperture (μSAXS). The positions and symmetries of the diffracted intensities in such μSAXS patterns reflect the symmetries of the short-range clusters within this volume [4]. The time correlations between patterns taken from the same volume in a time series show a characteristic decay (B) indicative of relaxations in the glass. In part C we show a map of the time correlation at an interval of 50 seconds obtained by scanning the same region of the specimen several times under the probe. We see that the dynamics of the system show local variations with a length scale comparable to the size of a co-ordination polyhedron. We compare this map to the magnitude of the 2-fold symmetry in the μSAXS pattern, mapped over the same region (D). We see that there is some correlation between areas with a large 2-fold symmetry and therefore pronounced centro-symmetry in the plane of the specimen and areas with a larger time correlation coefficient, and so greater similarities between configurations at different times. This indicates that local clusters in glasses with greater centro-symmetry have greater stablity and smaller amplitude dynamics, corroborating predictions [2].

While great progress has been made examining structure and dynamics in microcolloidal glasses with confocal optical microscopy, scanning mSAXS shows great promise for examining larger volume phenomena like shear localisation and understanding local structure in packings of smaller particles like nanoparticles and biomolecules. Analogous experiments could be performed on atomic glasses by employing electron nanodiffraction in the scanning/transmission electron microscope, uncovering universal behaviours.

This research was undertaken in part on the SAXS/WAXS beamline at the Australian Synchrotron, part of ANSTO.

[1] C P Royall and S R Williams, *Phys. Rep.* **560**, 1 - 75, (2015). "

[2] R Milkus and A Zaccone, *Phys. Rev. B,* **93**, 094204, (2016)

[3] A C Y Liu, R F Tabor, L Bourgeois, M D de Jonge, S T Mudie, and T C Petersen, *Phys. Rev. Lett.,* **116**, 205501 (2016)

[4] A C Y Liu, R F Tabor, L Bourgeois, M D de Jonge, S T Mudie and T C Petersen, *J. Stat. Mech. Theor. Expt.*, 054046, (2016).

[5] A C Y Liu, G R Lumpkin, T C Petersen, J Etheridge and L Bourgeois, *Acta Cryst. A*, **71**, 473-482, (2015)

[6] A C Y Liu, R F Tabor, M D de Jonge, S T Mudie, and T C Petersen*, Proc. Nat. Acad. Sci.,* **114**, 10344 - 10349, (2017)