ESRF	**Experiment title:** Dynamics and aging in non-Newtonian fluids under flow	**Experiment number**: SC-3292
Beamline: ID10A	**Date of experiment**: from: 23 Nov. 2011 to: 29 Nov. 2011	**Date of report**: 01 Sept. 2014
Shifts: 18	**Local contact(s)**: Yuriy Chushkin	*Received at ESRF:*

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Report:

Due to unforseen problems with the flow cell and analysis of data under shear flow we decided to change focus of the experiment.

We studied a model hard sphere suspension, comprising sterically stabilized PMMA particles suspended in decalin, at volume fractions between 0.55 and 0.61 The samples were filled into 1 mm diameter quartz capillaries for the measurements. Static SAXS pattern measured on a dilute ($\Phi < 0.01$) suspension at ID02 beamline was analysed to determine the structural properties of the sample. A Monte-Carlo based fitting method [1] has given the mean particle radus $R = 1024$ Å and polydispersity $\sigma_R/R = 0$ (see figure 1).

The XPCS measurements were performed using 8 keV X-ray beam, collimated to $10 \times 10\ \mu m^2$ size at the sample position. The scattering patterns were collected using the MAXIPIX detector, positioned about 2 m downsream of the sample.

The analysis of the Φ dependence of the long time diffusion coefficient D_l (see figure 2a), obtained from experimental data fits, is in line with the results found by Brambilla *et al*. [2]. $D_0/D_l(q_m)$ follows the Vogel-Fulcher-Tamman (VFT) form, $\frac{D_0}{D_l(q_m)} = \tau_\infty e^{\left[\frac{F}{(\Phi_0-\Phi)^\delta}\right]}$, with the fitted value of the critical volume fraction $\Phi_0 = 0.676$, matching the random close packing volume fraction for hard spheres of this polydispersity [3]. Along with the brakedown of the Segre-Pusey scaling [4] (figure 2b) and faster than exponential decay of the intermediate scattering function, observed for the most concentrated sample, this points towards the onset of collective motion in the vicinity of the glass transition and a dynamical slow down governed by reduction in free volume of the sample.

Multi speckle XPCS measurements have revealed the emergence of ergodicity restoring anomalous intermittent relaxation modes in the highest concentration samples (see the time-resolved correlation functions plotted in figure 3). We associate these phenomena with non thermal stress induced relaxations, which are a consequence of shear acting on the sample in the process of filling the capillaries used for the measurement.

These results have been accepted for publication in the Soft Matter journal.

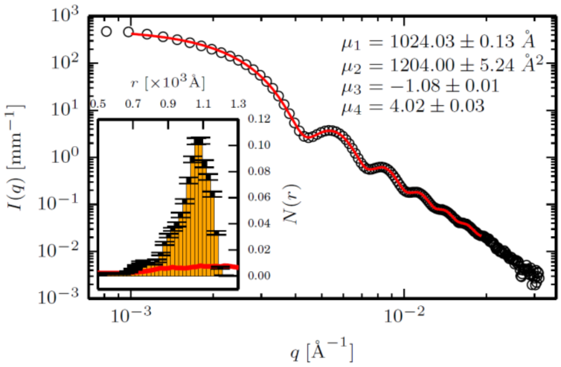

Figure 1 *SAXS data measured on a dilute PMMA suspension (open symbols), fitted with the MC simulation based method (solid red line). Inset: normalized, volume weigthed particle size distribution N(r). The solid red line is the minimum detection threshold. The values of the distribution moments are given in the main figure panel.*

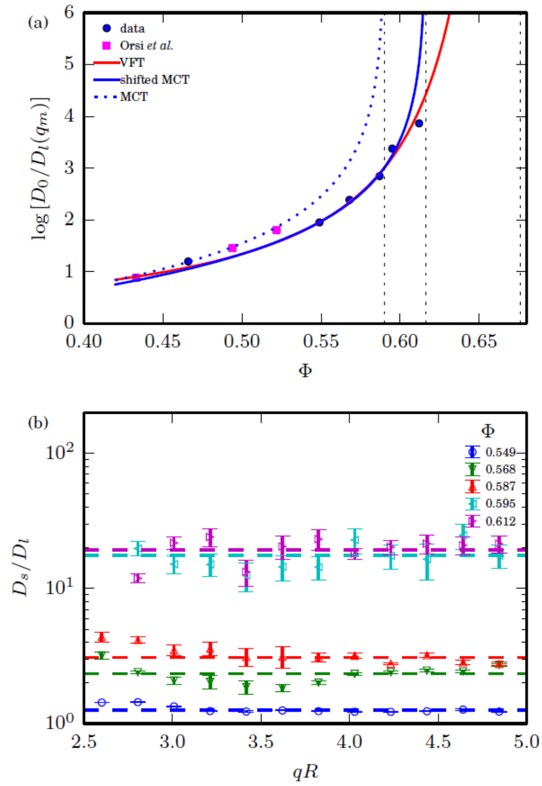

Figure 2 *(a) Experimentally obtained $D_0/D_l(q_m)$ (circles) compared to various models, as indicated in the legend. Additional data taken on the same system at lower concentrations (squares) are included in the fits and the figure.*

(b) D_S/D_l as a function of q for the concentrations probed. Dashed lines mark the average values. Only the suspension with smallest concentration (Φ = 54.9%) displays a q independent SP scaling of D_S and D_l.

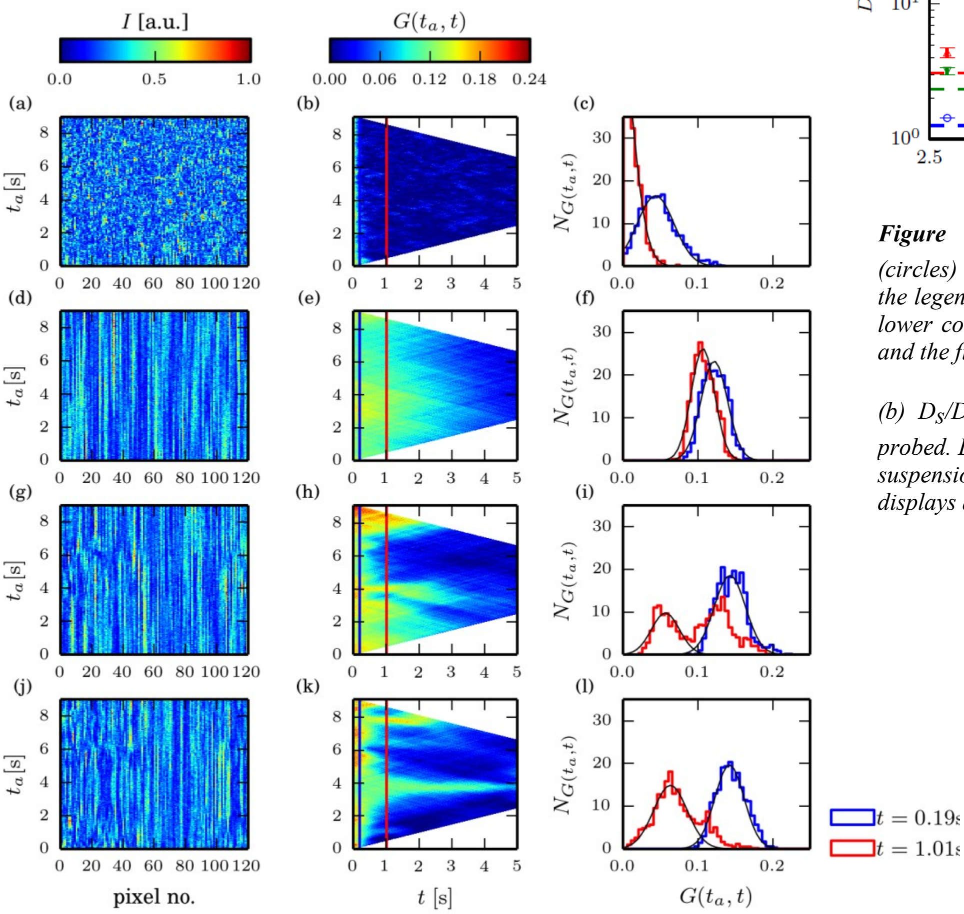

Figure 3 *Development of intermittent, heterogeneous dynamics in concentrated hard sphere suspensions. Left column: the intensity fluctuations as a function of time ("waterfall plot") for pixels at wave vectors near the $S(q)$ peak ($qR = 3.42$). Center column: plots of the time-resolved correlation function $G(q, t_a, t)$. Right column: histograms of G for two delay times t (marked with a blue and a red vertical line in $G(q, t_a, t)$). Panels a) - c): data for a sample with $\Phi \approx 0.57$. Panels d) - f): sample with $\Phi \approx 0.61$ after 30 min, g) - i) after 2.5h, j) - l) after 9h.*

References

[1] B.R. Pauw, J.S. Pedersen, S.Tardif, M. Takata and B.B. Iversen, *J.App. Cryst.*, 2013, 46, 365–371.
[2] G. Brambilla, D. El Masri, M. Pierno, L. Berthier, L. Cipelletti, G. Petekidis and A. B. Schofield, *Phys. Rev. Lett.*, 2009, 102, 085703.
[3] W. Schaertl and H. Sillescu, *Journal of Statistical Physics*, 1994, 77, 1007–1025.
[4] P.N. Segre and P. N. Pusey, *Phys. Rev. Lett.*, 1996, 77, 771–774.