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|                                                                                                                                                                                                                                  | <b>Experiment title:</b><br>Orientational order in interacting colloidal glasses | <b>Experiment number:</b><br>SC-3779 |
| <b>Beamline:</b>                                                                                                                                                                                                                 | <b>Date of experiment:</b><br>from: 23.10.2013 to: 29.10.2013                    | <b>Date of report:</b><br>18.09.2014 |
| <b>Shifts:</b>                                                                                                                                                                                                                   | <b>Local contact(s):</b><br>B. Ruta and F. Zontone                               | <i>Received at ESRF:</i>             |
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## Report:

One long-standing open question related to disordered materials is the nature and the origin of the glass transition. Colloidal suspensions are often used as model systems for studying this fundamental phenomenon. A key challenge is to unveil whether or not there is a structural change in the orientational bond order while the dynamics of the samples slow down at the glass transition.

In our previous experiment SC-3531 we studied colloidal hard spheres with different volume fractions by using X-ray cross correlation analysis (XCCA) [1]. With this technique we were able to observe the change of the local orientational order and we found indication for crystalline precursor structures [2].

In this experiment (SC-3779) we used charged stabilized colloidal suspensions. In contrast to hard spheres the phase transition does not only depend on the volume fraction but also on the particle charge. Therefore, the particle interaction in such systems can be tuned by adding salt to the colloidal dispersion [3].

In the framework of this experiment two different colloidal systems were studied: silica and polyacrylate colloidal particles. Both were dispersed in water with average particle radii of about 70 nm and 90 nm, respectively.

The experiment was performed in SAXS geometry at beamline ID10. The X-ray energy was set to 7.89 keV. The X-ray beam was subsequently collimated and focused to a final width of about  $10\ \mu\text{m} \times 10\ \mu\text{m}$  with a set of beam-defining slits. The samples were filled in quartz capillaries of 0.7 mm diameter which were sealed with glue and were mounted in the standard sample holder at ID10. The Maxipix 2x2 detector was placed at a detector-to-sample distance of 5.01 m. The pixel size of the Maxipix of  $55\ \mu\text{m} \times 55\ \mu\text{m}$  is comparable to the speckle size (around  $78\ \mu\text{m}$ ) that allows detecting the intensity fluctuations of the speckle.

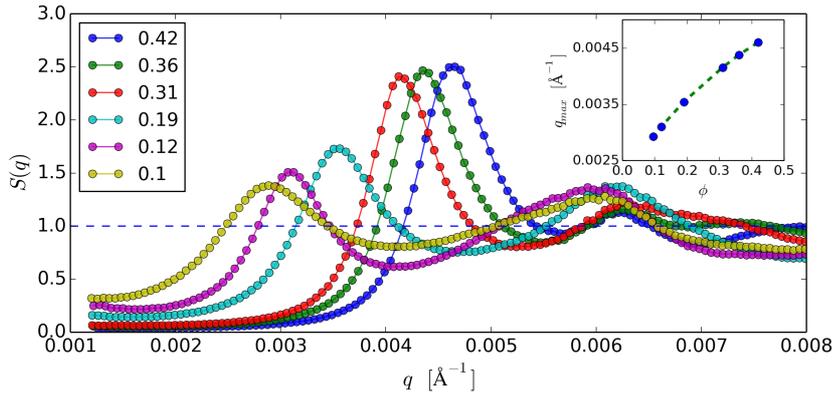


Fig.1. Static structure factor  $S(q)$  of the silica particles at different volume fractions  $\phi$  indicated in the legend. The inset shows the position of the first peak,  $q_{max}$ , of the structure factor as a function of the volume fraction  $\phi$ .

In a next step we measured the sample dynamics by XPCS to verify that the samples were slow enough, so that they were static within the exposure time. Such a constant behaviour is important for extracting information about local orientational order of the sample via XCCA. For XCCA, speckle patterns were taken at 1000-2000 different spots on the sample to avoid beam damage of the sample, at each spot at least 10 patterns were measured to check for consistency. The correlation analysis has been done according to the scheme in [1, 4, 5].

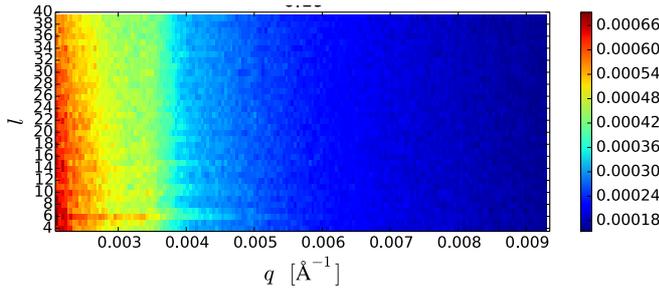


Fig.2. Fourier components  $|I_l|^2$  of the scattering intensity of a silica particles dispersion at a volume fraction of about 0.19. The color code represents the amplitude of  $|I_l|^2$

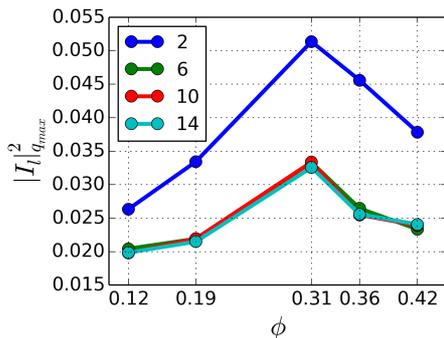


Fig.3. Evolution of the 2,6,10,14-Fourier coefficients as a function of the sample concentration.

The particle form-factors  $P(q)$  of the systems were measured from a dilute sample. The static structure factors  $S(q)$  were extracted for the different concentration of the samples by dividing the measured intensity by the particle form factor (fig. 1). The first peak of the static structure factor shifts to higher momentum transfers  $q$  with increasing particle concentration (fig.1(inset)), as expected for more concentrated systems.

As an example we show here calculated Fourier components  $|I_l|^2$  of the scattering intensity for the silica particles at a volume fraction of about 0.19 (fig. 2). We observe that the amplitude of each component is decreasing with  $q$  and the prevalent components are  $l = 2, 6, 10, 14$ . The value of the dominating Fourier components at the  $q_{max}$  position changes with the sample concentration (fig. 3). The components increase with the concentration up to a certain value, and then decrease. We believe, that the height of the Fourier components  $|I_l|^2$  correlate with the strength of the local symmetrical structure. However, for both silica and polyacrylate systems the data sets do not allow a definite statement due to a small amount of concentration values measured. In a subsequent experiment, we plan to extent our results by measuring samples with finer concentration steps.

## References:

- [1] P. Wochner et al. Proc. Natl. Acad. Sci. 106, 11511 (2009).
- [2] F. Lehmkuhler et al. Experimental Report SC-3531(2013), SC-3884(2014).
- [3] F. Westermeier et al. J. Chem. Phys. 137, 114504 (2012).
- [4] M.A. Schroer et al. Phys. Rev. E 90, 012309 (2014).
- [5] F. Lehmkuhler et al. J. Appl Cryst. 47, 1315 (2014).