



	<b>Experiment title:</b> Correlation of order beyond the first neighbour shell in complex liquids	<b>Experiment number:</b> SC-5105
<b>Beamline:</b> ID02	<b>Date of experiment:</b> from: 13.04.2021 to: 17.04.2021	<b>Date of report:</b> 06/12/2021
<b>Shifts:</b> 9	<b>Local contact(s):</b> Thomas Zinn	<i>Received at ESRF:</i>
<b>Names and affiliations of applicants</b> (* indicates experimentalists): Nele Striker*, Francesco Dallari*, Gerhard Grübel, and Felix Lehmkuhler* DESY, Hamburg Germany  Zhen Zhang and Walter Kob U Montpellier, France  Giulio Monaco U. Padova, Italy		

## Report:

In the proposed experiment, we wanted to probe the structure of different complex liquids beyond the first neighbour shell. Recently, a novel type of a four-point correlation function has been presented that probes the particle density in three dimensions from simulation snapshots of a dense Lennard-Jones liquid [1]. It was found that an intricate structural order exists in the simulated liquid given by alternating layers with icosahedral and dodecahedral symmetries that extends to surprising large distances (5-8 nearest neighbour shells). In order to measure such correlation, we proposed to perform an X-ray cross correlation analysis (XCCA) experiment in ultra-small angle scattering (USAXS) geometry examining correlations between different length scales (via different  $q$  values  $q_1$  and  $q_2$ ) to validate the predictions from simulations.

The experiment was performed at beamline ID02. Due to the Covid-19 pandemic, the experiment was performed remotely with users joining via videoconference. As samples we used five different batches of silica particles dispersed in water. These charge-stabilized systems show a rich phase behaviour covering liquid, crystal and glassy states [2]. The differences between the batches were basically the size of the particles and their dispersity, which results in small changes of the phase diagram – at higher dispersities crystallisation is suppressed and glass states can be achieved easily. Furthermore, due to the radii differences in the range of 40 nm and 100 nm, the particles have different charges which has as well impact on the phase behaviour. In general, the samples were measured at at least five different volume fractions between 0.01 and 0.30, thus covering liquid, supercooled liquid and glassy states [2]. We did not observe crystallisation during the measurement time thanks to the non-zero dispersity and pre-treatment (such as shaking) to avoid crystallisation or melt crystals. We decided not to measure hard sphere samples because they did not show significant structural correlations in a previous experiment at PETRA III as well as another experiment at ID10 [3].

We used a USAXS geometry with 20 m sample-detector-distance. For each sample we performed various X-ray photon correlation spectroscopy (XPCS) runs at different sample spots to measure the dynamics.

Therefore, we used the Eiger500k detector to achieve shortest time scales. Afterwards, the X-ray cross correlation analysis (XCCA) runs were performed. Here we changed to the Eiger4M detector to cover full q-rings necessary for XCCA. This was done by performing mesh scans on the sample and repeat the mesh several (up to 10) times which guarantees sufficient statistics for XCCA [4]. We could cover a q-range down to  $0.003 \text{ nm}^{-1}$ . This corresponds to length scales exceeding  $1 \mu\text{m}$  so that several next-neighbour distances can be covered in the  $q_1$ - $q_2$ -correlation analysis.

An example for the XPCS results from a dilute sample is shown in figure 1. At this low concentration the sample is in a liquid state and shows diffusive dynamics – highlighted by the  $q^2$  dependence of the relaxation rate and the KWW exponent of 1. Further analysis, with special emphasis on XCCA, is currently ongoing.

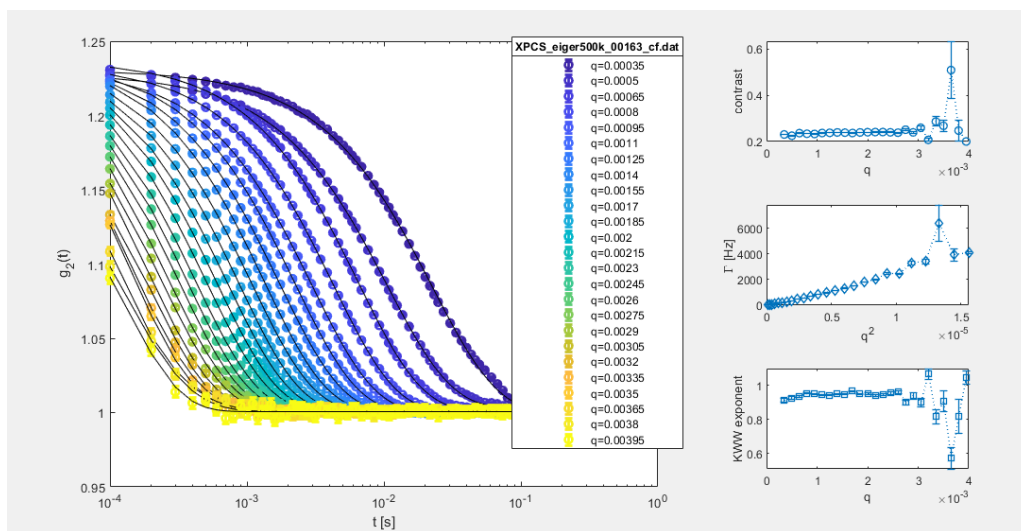


Figure 1: XPCS results for silica particles of 95 nm radius at a volume fraction of 0.01.

## References

- [1] Z. Zhang and W. Kob, PNAS 117, 14032 (2020).
- [2] E.B. Sirota et al. Phys. Rev. Lett. 62, 1524 (1989).
- [3] F. Lehmkuhler et al. Sci. Adv. 6, eabc5916 (2020).
- [4] F. Lehmkuhler et al. J. Appl. Cryst. 47, 1315 (2014).