

# Orientational order in colloidal suspensions studied by coherent x-rays

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Understanding the structure of liquids and glasses is one of the holy grails in condensed matter physics. However, the search for structural order is complicated because the ensemble averaged structure factors of liquids and glasses indicate the short range density-density correlation only. Molecular dynamics (MD) simulations showed that liquids and glasses develop a structural orientational order on a local scale [1]. Orientational order in glasses may remain from underlying crystallization processes or is suppressed by geometrical frustration via local symmetries, not extendable in 3D. Indeed, x-ray speckle patterns from colloidal glasses exhibit pronounced angular correlations [2].

In this experiment we studied the orientational order of mixtures of hard sphere colloidal suspensions via X-ray Cross Correlation Analysis (XCCA) [2, 3]. The experiment was performed at beamline P10 using the Small-Angle X-ray Scattering (SAXS) set-up with a sample-detector distance of 5 m. The detector was a Princeton Instrument CCD camera with a pixel size of  $20\mu\text{m}$ . The x-ray energy was chosen to 8 keV at a beam size of  $10 \times 10\mu\text{m}^2$ . As sample we used a mixture of poly(methyl methacrylate) (PMMA) spheres that are dispersed in decalin. The two constituents had a radius of  $r_1 = 128\text{ nm}$  and  $r_2 = 84\text{ nm}$  at polydispersities of  $\frac{\Delta r_1}{r_1} = 7.1\%$  and  $\frac{\Delta r_2}{r_2} = 9.2\%$ , respectively. The volume fraction was chosen around  $\phi = 0.56$  which corresponds to the glass phase. In total, we measured 13 different mixtures with particle number ratios between 0.2 and 5 (sample 1 : sample 2). First, we performed a quick X-ray Photon Correlation Spectroscopy (XPCS) run to measure the dynamics of the sample and thus confirming that the glass phase was set. Afterwards, we performed a long XCCA run by moving the sample to different position after a few patterns were taken. In this way we achieve different realisations of the sample.

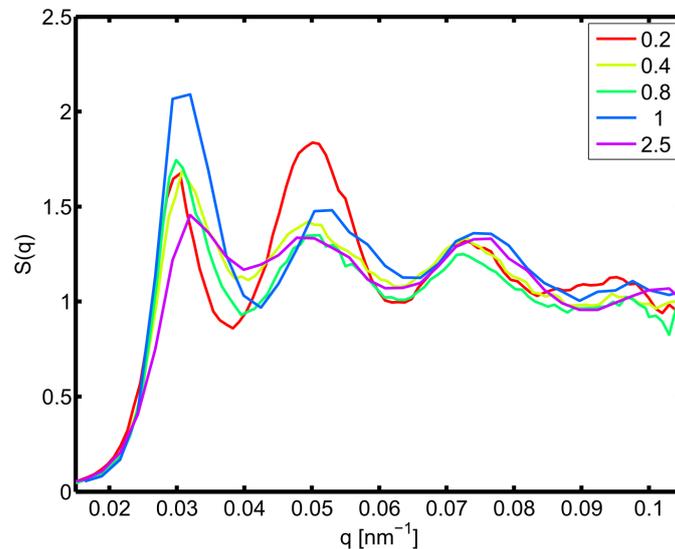


Figure 1: Static structure factor  $S(q)$  of selected samples. The number ratio is indicated.

Figure 1 shows a selection of static structure factors that were extracted from the scattering patterns for all samples. The highest maximum of  $S(q)$  can be found for a 1:1 ratio suggesting a higher averaged order than in the other samples. To identify the degree of orientational order, we calculate the correlation function [3]

$$\Psi(l, q) = \text{var}(I_l(q) / \langle I(q) \rangle_\omega), \quad (1)$$

where  $I_l(q)$  is the  $l$ th Fourier coefficient of the intensity  $I(q)$  with respect to the azimuth angle  $\omega$  of the two dimensional scattering pattern. This is shown in Fig. 2. Significant contributions can only be seen at very low  $q$  and  $l$ . However, after calculation of the order parameter

$$\xi(q) = \langle \Psi(l, q) \rangle_l \quad (2)$$

one can find a more detailed view on the angular correlations. There various maxima become visible at different  $q$  suggesting order on different length scales. In particular, the small maximum around  $0.025 \text{ nm}^{-1}$  can be connected to the typical distance between the large particles, while the second one around  $0.04 \text{ nm}^{-1}$  is in line with the typical small particle distance. These results will be compared with the results from the other samples and to simulation snapshots of spherical colloidal glasses.

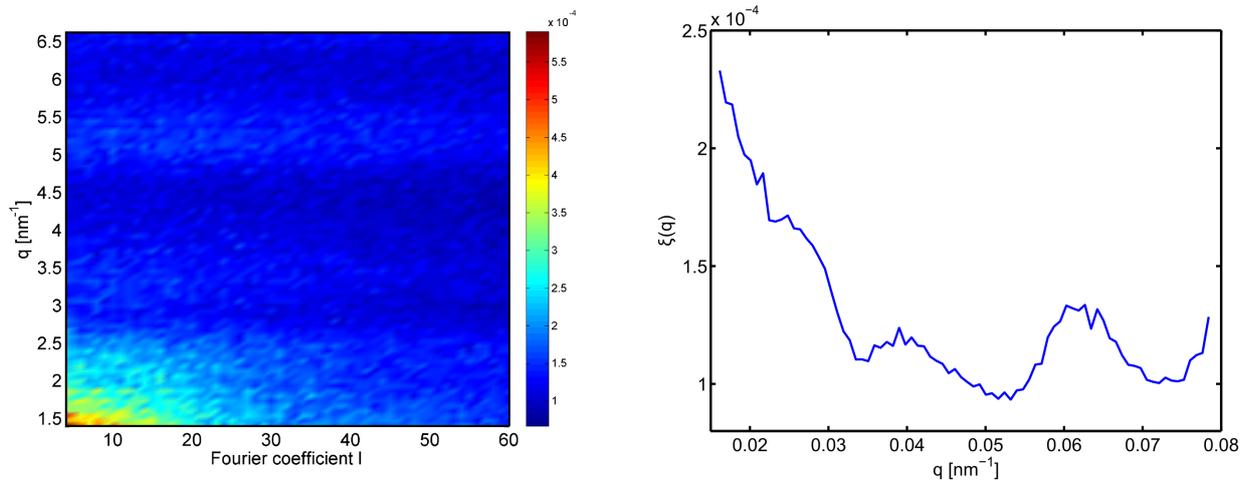


Figure 2: Left: Orientational order  $\Psi(l, q)$  of the 1:1 sample. Right: Order parameter  $\xi(q)$ .

## References

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