ESRF	**Experiment title:** Dynamic susceptibility measurements of colloidal suspension	**Experiment number**: HD-471
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Report:

Understanding the glass transition of supercooled liquids is a long-standing problem in condensed matter and statistical physics. In recent years, the research efforts have focused on the role of dynamic heterogeneity as the underlying reason for the increase in the viscosity of glass-forming liquids approaching the glass transition temperature T_G. Numerical simulations showed that near T_G the dynamics becomes increasingly correlated in space. One of the basic problems is to find an observable that allows to measure and quantify such a correlation length. A candidate for that is the dynamic four-point susceptibility χ_4 based on the four-point density correlator G_4

$$G_4(\vec{r},t) = \langle p(0,0)p(0,t)p(\vec{r},0)p(\vec{r},t)\rangle - \langle p(0,0)p(0,t)\rangle\langle p(\vec{r},0)p(\vec{r},t)\rangle.$$

Theory predicts a rich time structure of $\chi_4(\tau) \propto \int d\vec{r}\, G_4(\vec{r},\tau)$ with notable differences between common glass formation models in the time sectors of α- and β-relaxations. χ_4 can be measured in X-ray photon correlation spectroscopy (XPCS) experiments using colloidal particles in highly viscous solvents. Typical features are a peak structure of χ_4 and the typical hallmarks of heterogeneous dynamics as e.g. ballistic motion.

We have investigated the χ_4 of supercooled colloidal suspensions with respect to colloidal particle sizes and colloidal concentrations. In this experiment, we measured different samples of a concentration series (0.5–5% volume fraction) of stabilized SiO_2 colloidal particles (radius ≈ 48nm and 81 nm) suspended in the molecular glass former dibutyl-phtalate (DBP) (T_G = 186 K). We recorded time-series of coherent small angle x-ray scattering (SAXS) patterns with a Andor CCD area detector at different temperatures around the molecular glass

transition of the solvent. Measurements have been performed at different temperatures covering T = 187–298 K. We applied time-dependent higher order correlation functions to the data and extracted the temporal-resolved information by the instantaneous degree of intensity correlation between pairs of speckle images, C_I, as a function of absolute time t and delay τ between images given by

$$C_I(q,t,\tau) = \frac{\langle I_p(t) I_p(t+\tau) \rangle_{p \in q}}{\langle I_p(t) \rangle_{p \in q} \langle I_p(t+\tau) \rangle_{p \in q}} - 1,$$

where $I_p(t)$ is the scattered intensity at pixel p and time t. The brackets denote the average over circular regions of pixels corresponding to a mean value of equivalent q. χ_4 is then extracted by calculating the variance of C_I.

A first analysis of the data (Figure 1) shows that the particle size seems to affect the shape of χ_4 more strongly than the colloid concentration. In comparison to other measurements, multiple relaxation times (τ) become visible in χ_4 only at the smallest colloid-particle size (radius 48nm). It is tempting to interpret such an observation as fingerprint of the size of cooperative rearranging regions. Such a finding may give direct access to temperature dependent size limits of the cooperative rearranging regions, which would advance the understanding of glass physics considerable.

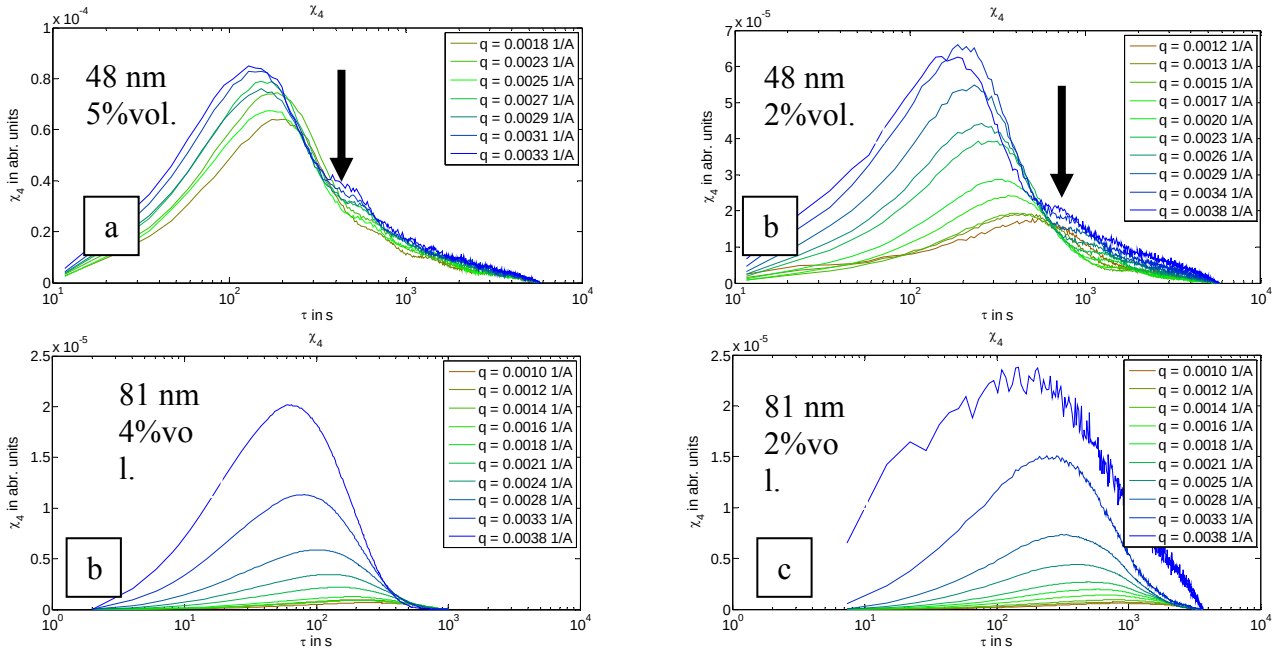

Figure 1: Dynamic susceptibility χ_4 of SiO_2 particles solved in DBP at T = 185 K ([a-b] radius = 48nm, [c-d] radius=81nm, respectively).