ESRF	**Experiment title:** Dynamic Susceptibility Measurements of Colloidal Suspensions – a follow up	**Experiment number:** HD 585
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Report:

Although glasses are very common in everyday life, the transition from supercooled liquids to the glassy state is one of the mysteries in condensed matter physics [1]. Besides a slowing down of the dynamics by orders of magnitude, relaxation phenomena become non-exponential pointing to a broad distribution of the relaxation times and existence of dynamical heterogeneities upon approaching the glass transition. This is suggested to be closely connected to spatial heterogeneities where different local arrangements can be found within the liquid showing a variety of relaxation times. These dynamical heterogeneities can be accessed by higher-order correlation functions [2]. In particular, using coherent x-rays as probe heterogeneous dynamics can be studied via two-time correlations that are determined from X-ray Photon Correlation Spectroscopy (XPCS) experiments [3]. In this experiment we studied the dynamics of polypropylene glycol (PPG) with the help of colloidal tracer particles by means of XPCS in a temperature range from room temperature to the glass transition temperature of $T_g = 205$ K [4].

The XPCS experiments were performed at beamline ID10 in SAXS geometry using the MAXIPIX 2x2 detector. The photon energy was set to 8 keV at a beam size of 10x10 μm^2. The samples (silica spheres of 70nm radius dispersed in PPG, volume fraction approximately 1 %) were filled in quartz capillaries of 1mm diameter that were vacuum sealed afterwards. These capillaries ware placed in a cryo-SAXS chamber that was cooled by gaseous nitrogen flow [5]. First, SAXS patterns were taken at room temperature (295 K). Afterwards, the temperature was decreased in several steps (typically 5-10 K at high temperature and 1-3 K below 230 K) down to the glass transition temperature of PPG of 205 K. At each step, an

XPCS run was taken with adapted exposure and delay times in order to cover the dynamics of the sample at the given temperature.

The obtained intensity correlation functions g_2 were fitted by the model $g_2(q,t) = \beta\, exp(-2[\Gamma(q)]t^{\gamma})+1$, with the speckle contrast β, the decay rate Γ, lag time t and the stretching exponent γ. In addition, the q-dependence of Γ was modeled by $\Gamma(q) = Dq^p$. In case of p=2, the particles undergo diffusion, while p=1 is connected to ballistic motion. The resulting exponents are shown in Figure 1 as function of temperature.

Based on these results, three temperature regions can be defined. At high temperatures (region B) Brownian motion of the particles is observed ($p \approx 2, \gamma \approx 1$). Close to T_g(region A), the dynamics is hyperdiffusive and ballistic ($p \approx 1, \gamma \approx 2$). At intermediate temperatures around 1.12 T_g we observed an intermediate region (AB) with $p \approx 1.5, \gamma \approx 1.5$. Moreover, γ was found to be q-dependent in this region. Similar results have been reported in previous experiments on different glass formers [6] and were suggested to be originated by cooperative dynamics at low temperatures.

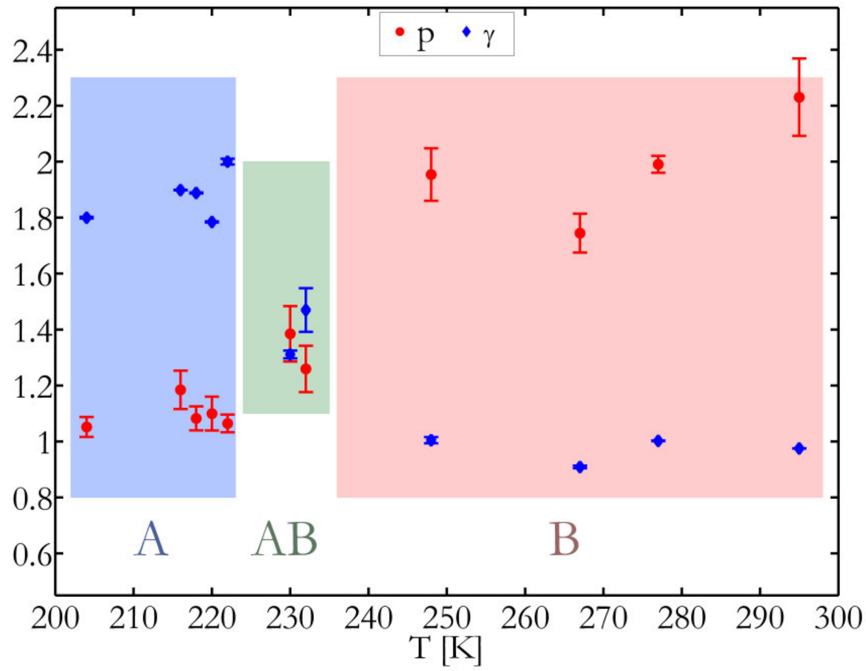

Figure 1: Temperature dependence of the exponents γ and p [4].

In order to gain deeper insight into this dynamics cross-over, we analyzed higher-order correlations of the scattering data by calculating the four-point dynamic susceptibility χ_T [3]. We found a dominant peaking at the inflection point of the g_2-functions. With decreasing temperature the peak height increases dramatically when reaching the intermediate temperature regime (region AB). This shows that the dynamics becomes increasingly heterogeneous around 1.12, indicating the existence and growth of rearranging cooperative regions [1].

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