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<tr><td rowspan="1">ESRF</td><td>Experiment title: Spatial investigation of dynamical heterogeneities in soft attractive and repulsive glasses</td><td>Experiment number:<br>SC-4541</td></tr>
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## Report:

The aim of the present experiment was to characterize the dynamical properties of a colloidal system close to the glass transition. More precisely we wanted to obtain the q-dependence of the dynamical susceptibility ($\chi_4$) for colloidal glasses composed by hard spheres with particle-particle interaction given by either a repulsive or an attractive force.

In fact, theoretical studies [1,2] predict a (q,t) dependence of the $\chi_4$ for these prototypical systems, depicting two qualitatively different scenarios depending on the details of the inter particle interaction (i.e. repulsive forces or attractive forces). With this fact in mind, the system chosen for our experiment was a colloidal glass of silica nanoparicles dispersed in a near critical binary mixture of water and lutidine 2,6. This peculiar system offers the possibility to tune the inter-particle interaction with temperature, however the temperature range in which this control is made possible is rather limited and very close to a phase separation, thus a very high thermal stability and homogeneity across the sample are necessary in order to exploit successfully this property.

The $\chi_4$ has been measured using two different independent approaches (both of them exploitable even in non-equilibrated samples) to obtain a full control of the results. In the first approach the $\chi_4$ has been obtained as the variance of the experimental contrast in a standard g2(t) measurement, following [3], the drawbacks of this method is the necessity to perform an heavy processing of the data in order rule out spurious contributions due to i.e. statistical fluctuations and aging of the sample. In addition, the $\chi_4$ can be also measured exploiting a combination of homodyne and heterodyne X-ray photon correlation spectroscopy (XPCS) following the scheme reported in [4], which in principle lighten the computational efforts needed to obtain the required physical quantity.

We carried out small angle X-ray scattering experiments (SAXS) with photon energy of 8.1KeV, on two different sets of colloidal suspensions, one composed by nanoparticles with diameter of 450 nm and the other composed by nanoparticles with diameter of 100 nm. In both cases we investigated concentrations ranging from volume fractions of approximately 50%, (corresponding to a very slow dynamics, close to the arrested condition ) to very diluted samples.

Thanks to the high density difference between the silica particles and the binary mixture of water-lutidine we obtained good quality SAXS data see fig 1(a) . In order to rule out potential effects due to beam damage we systematically performed measurements at different beam fluxes on the same samples. No presence of damage has been observed on the long run for attenuations of 10^-4 or greater.
The heterodyne detection was realized with small quantities of a silica xerogel placed immediately upstream the sample. We observed that the xerogel can produce a nearly uniform static field on the detector area, which is an advantageous feature for a local oscillator. However, it turned out that for q-values corresponding to the first peak of the structure factor, the local oscillator signal had an intensity comparable to the signal coming from the sample. This is complicating the analysis of the data, which is in progress.
Due to technical limitations, it wasn't possible to investigate the dynamics in both the attractive and repulsive regimes during our beamtime, more in detail it wasn't possible to establish an homogeneous temperature across the sample with the sample holders available at that time; thus only the repulsive glass has been investigated.
At the moment, we succeeded in obtaining good results for the $\chi_4$ with the method discussed in [3], in fig.1(b) is shown an example for a colloidal glass of 100 nm particle at volume fraction of approximately 40%.
We are now working on the comparison between the q-dependence for $\chi_4$ that we have obtained for samples with different volume fractions of silica nanoparticles and the theoretical prediction in Ref. [1].

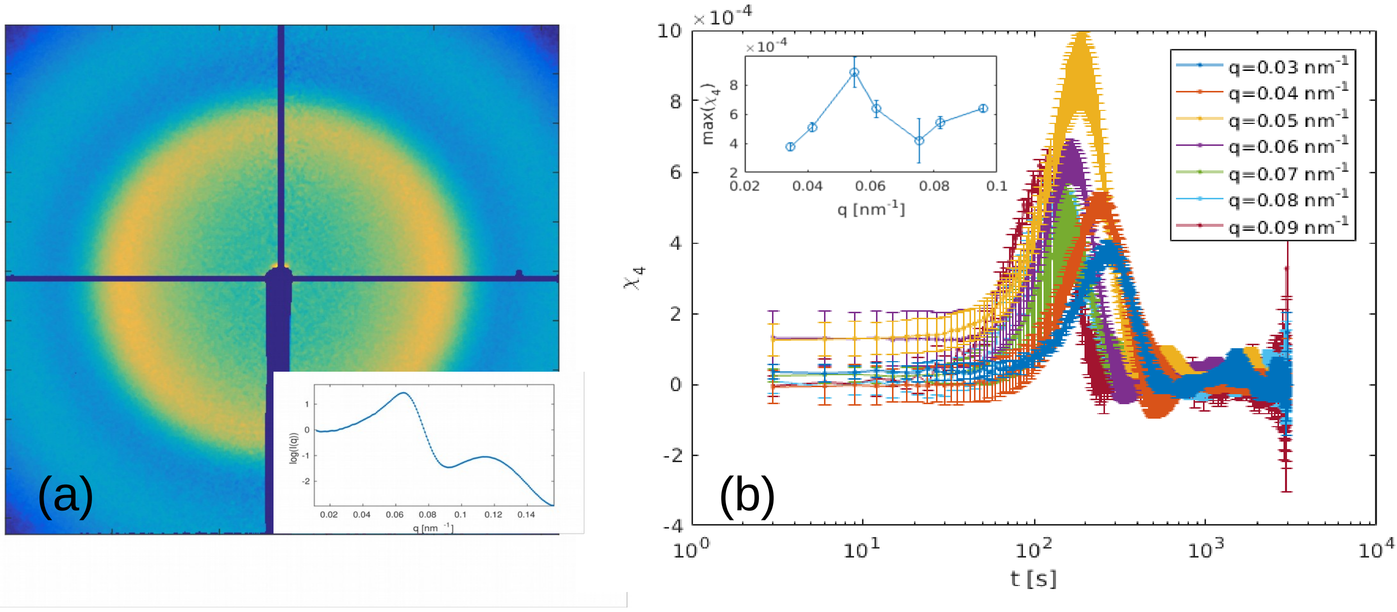


*Figure 1(a): Signal from a sample of 100 nm silica particles in water-lutidine; inset: average intensity as a function of q. Figure 1(b) extrapolated values of the dynamical susceptibilities for an aged sample of 100 nm colloidal glass; inset : peak values of the $\chi_4$ as a function of q.*

## References:


[1] P. Charbonneau and D. R. Reichman *Phys. Rev. Lett*. 99, 135701 (2007)
[2] Xinhui Lu, S. G. J. Mochrie, S. Narayanan, A. R. Sandy, M. Sprung *Phys. Rev. Lett*. 100, 045701 (2008)
[3] V. Trappe, E. Pitard, L. Ramos, A. Robert, H. Bissig, and L. Cipelletti *Phys. Rev. E* 76, 051404 (2007)
[4]C. Maggi, R. Di Leonardo, G. Ruocco, J. C. Dyre *Phys. Rev. Lett.* 109, 097401 (2012)