ESRF	**Experiment title:** Anisotropic aging in glassy Ferrofluids	**Experiment number**: SC-2095
Beamline: ID10C	**Date of experiment**: from: 14/02/2007 to: 20/02/2007	**Date of report**: 20/03/2007
Shifts: 18	**Local contact(s)**: Dr. Anders MADSEN	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):

Pr. Régine Perzynski[1]*, Ph.D. Elie Wandersman[1]*, Dr. Emmanuelle Dubois[1]*,

Dr. Vincent Dupuis[1]*, Dr. Aymeric Robert[2]*

(1) Lab. LI2C, UPMC, 4 Place Jussieu, case 51, F75252 Paris Cedex 05, France

(2) SLAC, Stanford University 2575 Sand Hill road, MS43A, CA94025 Menlo Park USA

Report :

The aim of the proposal was to investigate the slow dynamics of glassy magnetic colloidal dispersions consisting of charge-stabilized spherical magnetic nanoparticles (γ-Fe_2O_3) dispersed in water in various repulsive situations in zero field and under an applied field. Indeed in these glass-forming samples, the interaction potential can be tuned through several physico-chemical parameters, namely the nanoparticle size, the ionic strength and the osmotic pressure. Moreover applying a magnetic field introduces an anisotropy of interaction inside the system, which induces an anisotropy of aging properties.

Therefore we used the 2D-X-ray Photon Correlation Spectroscopy (2D-XPCS) capabilities of the ID10C Troïka beamline. The sample was placed in a specific sample environment (borrowed from ID02) allowing applying a permanent homogeneous magnetic field perpendicular to the X-ray beam (up to 0.4 T). We need to use X-rays below the Fe-absorption K-edge (at 7.112keV) with a large enough X-ray intensity. This was achieved in our former experiments with 2 undulators tuned at 7.06keV and a beam of section 40*40μm^2. We thus asked in our proposal for these same conditions (or for 3 undulators properly tuned).

Because of problems with the beam-line schedule, we have been proposed a setting different from the one we have asked for. We obtained only one undulator tuned at the proper energy; the flux was then far too low for the realization of the experiment. The local contact proposed to change the optical setting of the beam line with X-ray lenses in order to increase the incident flux, at the price of the degree of coherence of the beam. This last point has been compensated by a reduction of the beam size inside the sample (down to 5*5μm^2). So we have performed the experiments with a larger energy density than expected, and achieved a different ensemble averaging because of the serious reduction of the beam size.

A direct illumination deep depleted CCD, performing in single-photon-counting mode with pixels size of 20*20 μm^2 was requested to record the time-resolved speckle patterns scattered by the coherently illuminated sample. The CCD camera that we have been initially using had burns and we have thought that it would affect our data. We thus took the time to verify that indeed a change of camera had a major influence on our data quality.

Three different samples (with same particle size and ionic strength but a different osmotic pressure, resulting in various concentrations and thus various states of interparticle repulsion) were investigated without magnetic field. Due to the experimental difficulties encountered at the begining of the experiment we have studied only one sample of them under magnetic field. A conventional (i.e non-coherent) SAXS measurement was first performed on a dilute sample in order to get the form factor of the probed nanoparticles. Time-resolved speckle patterns were recorded over 11 hours (being limited to 12h due to the refill of the machine with almost 1h needed to properly align the sample and record dark files for correct background substraction of the CCD frames). Each frame was taken over a typical acquisition time of 10s (+2.5s for readout), time required to get sufficient statistics on each CCD frame to perform 2D-XPCS calculation with an acceptable signal to noise ratio.

We recovered with the new samples (and the new setting) general features observed in our former experiments : the correlation peak related to the static structure peak of this glassy system located around Q=0.045 $Å^{-1}$, isotropic speckle patterns in zero field, anisotropic under applied field, obvious indications of aging in the dynamics from the preliminary data analysis. However the new setting (i.e. with a focussed beam) of the beam line gave us an optical signal with much larger dynamical fluctuations than previously. We suspect that this is closely related to the difference in ensemble averaging realized with the two kinds of setting (i.e. different beam size at the sample). It gave us an idea for our new proposal, namely to ***analyze quantitatively the temporal heterogeneities of our samples as a function of the coherently illuminated volume***. The amplitude of the fluctations observed here is clearly decreasing with the nanoparticle concentration (see fig. 1). We thus performed several tests to check the feasability of the Time Resolved Correlation analysis of these fluctuations.

- We have checked in a closing/opening process of the X-rays beam that the incident energy density of X-rays was not inducing a rejuvenation of our aging sample,
- We have recorded time-resolved patterns with one sample (fig 1 right) during 12h more, without moving the capillary containing the sample inside the chamber, in order to check that we could obtain reliable data for a Time-Resolved Correlation analysis at larger age.
- This aging cannot be realized outside the beam as we have checked that taking out (delicately) the sample from its position and putting it back for counting, rejuvenates it.

Moreover the mecanical vibrations induced by the set-up borrowed from ID02 which produces a magnetic field of varying amplitude (by moving large permanent magnets) forbid us to study any rejuvenation induced by a pulse of magnetic field (as it was proposed in the present proposal). A new sample environment has to be imagined and built up in the future.

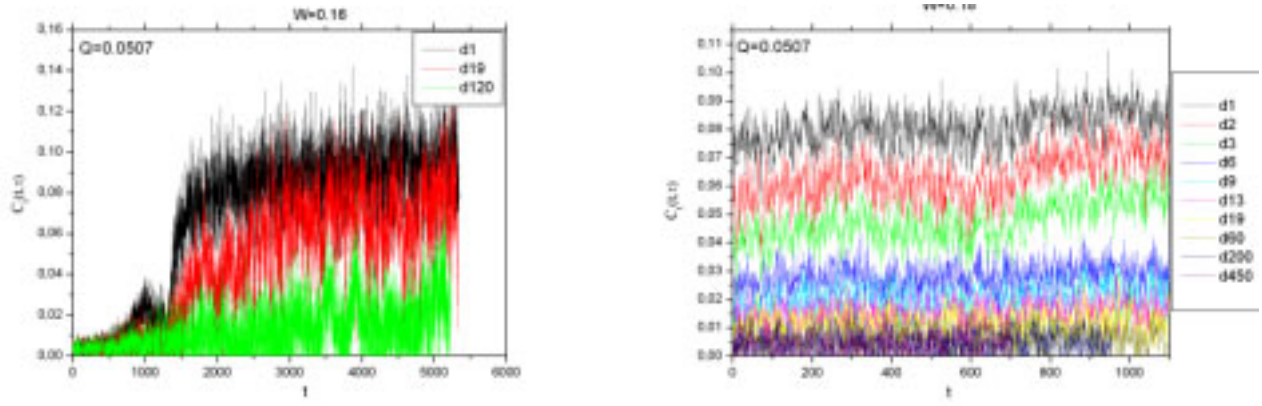

Figure 1 : Degree of correlation C_I betwen pairs of isotropic speckle patterns (measured without magnetic field) at Q=0.0507$Å^{-1}$ at times t and t+τ as a function of t (t and τ are expressed in terms of image number) (left) : Sample W=0.16; (Righ) : Sample W=0.18 (being already in place for 12h). W (a.u.) is an increasing function of the nanoparticle concentration inside the sample.