ESRF	**Experiment title:** Glassy dynamics of magnetic nanoparticles in a supercooled liquid	**Experiment number:** Sc2823
Beamline: ID10	**Date of experiment:** from: 9/12/09 to: 14/12/09	**Date of report:** 10/02/09
Shifts: 17	**Local contact(s):** Y. Chushkin	*Received at ESRF:*
Names and affiliations of applicants (* indicates experimentalists): E. Dubois, R. Perzynski, Laboratoire PECSA, UPMC, 4 place Jussieu, case 51, 75005, Paris		

Report:

The aim of the experiment was to measure the dynamics of magnetic nanoparticles, without and with field, in a supercooled solvent for concentrated dispersions that allowed to obtain enough intensity. The samples studied were based on maghemite nanoparticles ($\gamma - Fe_2O_3$) dispersed in dibutylphtalate using a surfactant, which was the only system, among the seven tested in our first experiment in 2008 (see rep. SC2337), that resisted to beam damage.

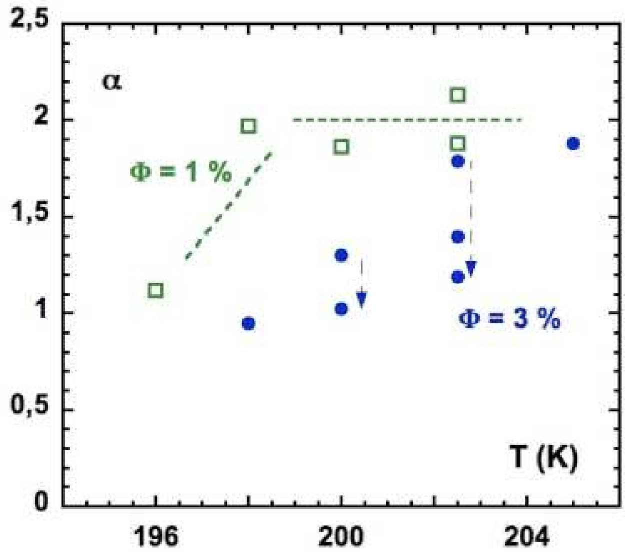
The experiment has been performed on ID10C with X-rays at 7.05 keV ($\lambda = 1.75\mathring{A}^{-1}$), lenses and a 15x15 μm beamsize. The samples were placed in capillaries in the low temperature SAXS chamber, with capabilities of applying external magnetic field H perpendicular to the beam propagation. Temperature stability was better than 0.01K. The scattered intensity was measured by direct illumination CCD (Andor) with the pixel size 13x13 μm . The correlation function were obtained by processing the CCD images using multispeckle technique.

During this experiment, we discovered that the beamline had been very highly improved so that the signal was much higher than during the previous experiment. Indeed, with our tests, only concentrated samples with a volume fraction Φ around 12-15% were measurable, with images of several seconds. In december 2009, we were able to measure such concentrated samples but also samples much more dilute with Φ=3% and 1% with images around 1 s or less, which opened new perspectives.

In the present experiment, the concentrated sample (Φ=14%) gave results similar to the first tests : the dynamics is anisotropic at low temperature (around 200K) without magnetic field. This was previously attributed to the sedimentation of concentrated droplets of concentrated magnetic fluid resulting from a phase separation at low temperature.

This has been confirmed by the new experiments on more dilute samples. Indeed, for $\Phi = 3\%$ and 1%, the dynamics is isotropic without field in the temperature range

explored (205K - 196 K). Under field (H= 1200 Oe), only the sample with Φ=1% has been measured : its dynamics remains isotropic at 202.5K and becomes anisotropic at 196K, with a dynamics faster in the vertical direction (perpendicular to $\vec{H}$) and an oscillating correlation function (see Figure 1 right). This corresponds to the formation of droplets of concentrated phase induced by H, and explains the behaviour of the concentrated samples for which the droplets already form without H at low T.
With these dilute samples, we are able to follow the dynamics in the range 196K $\leq T \leq$ 205K. On this range, the inverse characteristic time Γ of the correlation functions shifts from a Q^{α} variation with $\alpha = 2$ at high T's towards $\alpha = 1$ at low T's (see Figure 1 left), which is consistent with previous measurements by XPCS on bigger silica colloids [1]. Moreover, the χ_4 determinations support the existence of cooperative processes at the lower T's.
For these two samples and the solvent, a value of Tg = 182 K is measured by DSC, however the behaviours are shifted in the 3% sample : the dynamics is slower at a given T than for 1%. It means that the interactions between particles already modify the behaviour at 3%, which presents slow and ballistic-like dynamics together with aging at $T \leq$ 202.5K.
Under field, the dynamics of the more dilute sample at high T is nearly isotropic, showing that the interparticle interactions are low enough and do not perturb the dynamics. At lower T's, when the dynamics becomes anisotropic due to the droplets, the dynamics parallel to H, not perturbed by sedimentation, is slower than with H = 0 (see Figure 1 right), which confirms that the dynamics then probed contains a contribution from the internal dynamics of the droplets, which are more concentrated than 1%.



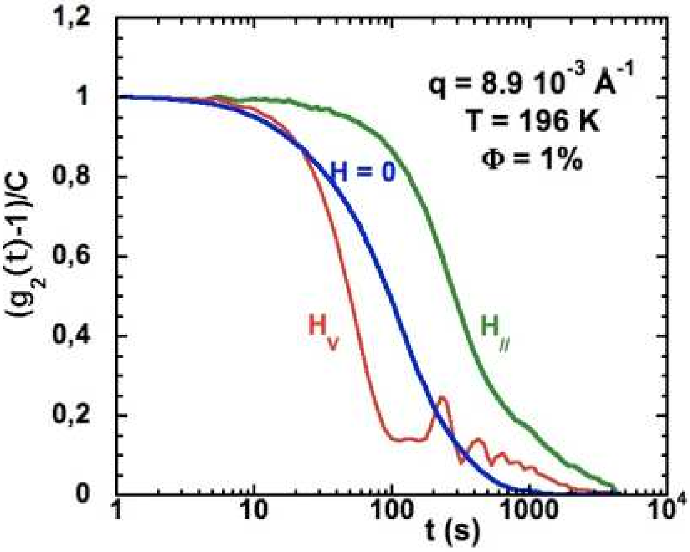


Fig.1 : Left : Exponent α determined from the variation $\Gamma = \tau^{-1} = Q^{\alpha}$ for the two dilute samples versus T; Right : Correlation functions for dilute sample with H=0 and H=1200 Oe at 196 K.

[1] C. Caronna et al., PRL 100, 055702 (2008); H. Guo et al. PRL 102, 075702 (2009)