ESRF	**Experiment title:** The dynamics of colloids at liquid-liquid interfaces	**Experiment number:** SC2084
Beamline: Troika ID10	**Date of experiment:** from: 20/09/06 to: 25/09/06	**Date of report:** 28/02/07
Shifts: 18	**Local contact(s):** Dr. Aymeric Robert	*Received at ESRF:*

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Report:

The aging of soft materials is scientifically very intriguing and far from understood. Our previous studies of dense oil-in-water emulsions stabilized by colloidal silica demonstrated that these systems show complex aging behaviour. As a consequence we carried out this study with careful control of when the emulsions were formed.
The systems under investigation are solid-stabilised emulsions where the droplets have creamed to the top of the sample. The stabilising agent are monodisperse 0.5μm diameter silica spheres prepared after the Stöber method. On adding appropriate amounts of salt these particles become irreversibly trapped at the water-oil interfaces. The oil used here is dodecane and to obtain emulsions the system is mechanically agitated. To investigate the changing dynamics of our systems systematically we have now carried out a set of experiments where we observe our samples over an extended period of time only varying the volume fraction of particles while keeping all other parameters the same. By changing the volume fraction of the added particles we can reproducibly vary the size of the resulting emulsion droplets. The size of the droplets, ξ varies inversely with the volume fraction of the particles Φ_{vol}. $\xi \propto \frac{r}{\Phi_{vol}}$[1], where r is the radius of the stabilising particles. Both the bouyancy force, that leads to creaming, and the interfacial curvature will vary systematically with the average droplet size. We are exploring how these parameters control the aging behaviour.
Due to the high opacity of the samples conventional light scattering is unsuitable to study this system. Furthermore it is necessary to have a large q-range with very good statistics. This can be achieved with the XPCS setup on the Troika beamline. In the previous experiment we have established that the water-dodecane system with 0.5μm

diameter silica particles produces scattering data suitable for observing the network aging.

During this experiment we were able to collect data in a q-range from $1 \times 10^{-3}\mathring{A}^{-1}$ to $10 \times 10^{-3}\mathring{A}^{-1}$ for four different volume fractions observing the samples from a few minutes after preparation up to over 100 hours. As before slow speckle fluctuations were visible by eye and normalised intensity-intensity correlation functions were obtained by using a software multi-tau, multi-Q correlator[2] to process the collected data.

The analysis is currently in progress as part of Eva M Herzig's PhD thesis and will be carried out over the next few months. The initial evaluation of the data shows how the g_2-1 curves for an 8h old emulsion initially still drop off to zero while at later times (70h) a plateau is reached (Fig.1). This indicates some form of arrest of the system and is expected since the walls have been shown to solidify[3]. In the upcoming analysis we will focus on the relaxation of the normalised intensity - intenstiy correlation function g_2. For glassy soft matter systems this follows $exp(-(\frac{\tau}{\tau_c})^{\beta})$ where β values can deviate from 1.0[4]. The variation of τ_c with q provides information on how the particles rearrange within the system. We are interested to see if the behaviour of β and τ_c varies with the different stages in the aging of our emulsions therefore shedding more light on the mysterious aging of these systems. The aging behaviour appears to be associated with the droplets becoming faceted and fusing together due to bouyancy. Our preliminary analysis suggests that emulsions with higher curvature droplets show more dramatic changes with emulsion age.

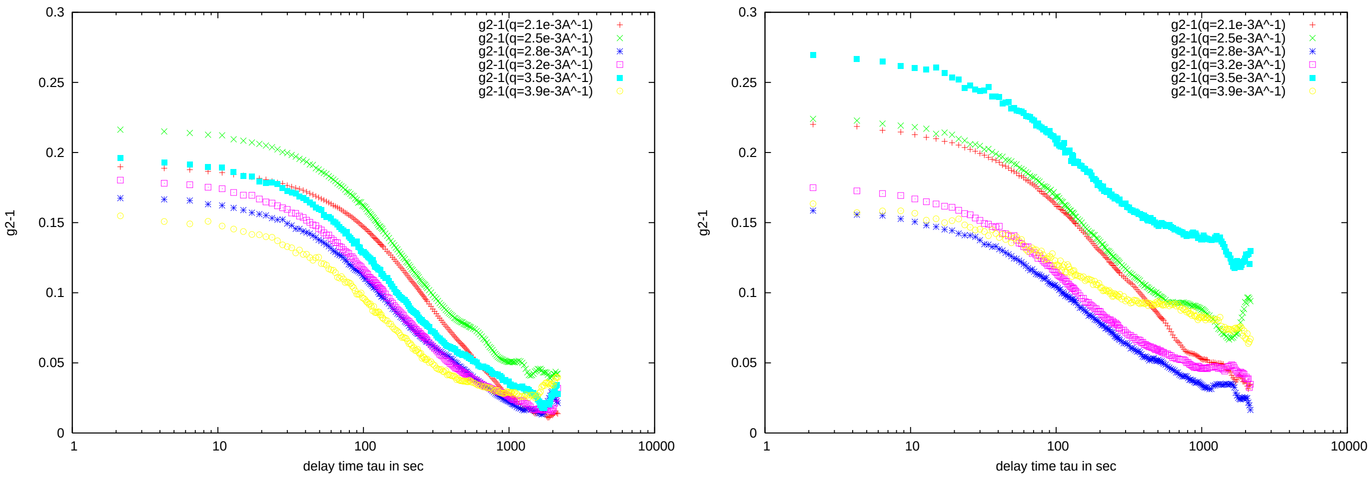

Figure 1. Example of calculated correlation functions at a range of wave vectors for a water/dodecane emulsion stabilised with 500nm diameter silica particles 8h after preparation (left) and 70h after preparation (right).

Luca Cipelletti kindly agreed to let us use his code for the analysis once again and supplied us with additional extensions to test for intermittency. At all stages of the ongoing experiments we received once again outstanding support and advice from Aymeric Robert. We have also received assistance from Yuriy Chushkin.

References

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[2] Cipelletti et al., Rev. Sci. Inst., **70**, 3214 (1999)
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