ESRF	**Experiment title:** **Short-range dynamics of nematic suspension**	**Experiment number**: SC-2442
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Report:

The purpose of this experiment was to study the low-frequency dynamics of a nematic phase of goethite nanoparticles. X-ray photon correlation spectroscopy (XPCS) is a unique tool for the study of such concentrated colloidal suspensions, as it can probe the dynamics down to wavelengths of the order of the interparticle separation.

Goethite is an iron oxyhydroxide with the general formula FeOOH. Following a well-established synthesis, it can form rodlike, polydisperse nanoparticles[1] with an average size of $150\times25\times10\ nm^3$. At low volume fractions, aqueous suspensions of goethite nanoparticles are isotropic. At concentrations above 6.7 vol%, the suspensions become nematic. Moreover, the suspensions have interesting magnetic properties. The nanoparticles align with their long axes parallel to a low-strength magnetic field (B<250 mT), and perpendicular to a high-strength magnetic field (B>250 mT). Thus, a nematic phase can be aligned by such a field and form a monodomain.

For this experiment, the goethite nanoparticles were resuspended in a mixture of propane-1,3-diol and water, which has a significantly higher viscosity than pure water, and, more importantly, a much lower freezing point. Relaxation rates were obtained at 228K, temperature at which the viscosity of the solvent is about 1,000 times higher than that of pure water. The increased viscosity serves to slow down the dynamics of the particles so that the XPCS experiment becomes feasible.

Isotropic suspensions at various volume fractions were put in 50μm flat optical capillaries. The relaxation rates as a function of scattering vector q were measured using the Medipix camera. For each sample, 10,000 speckle patterns were recorded with a 0.01 s time delay between them. The intensity correlation functions g(t, q) were calculated for each pixel separately, and then averaged for pixels at the same q. A nematic suspension at 6.7 vol% was also measured. It was first oriented by the application of a weak magnetic field (B = 150mT), and the correlation functions were obtained in the direction perpendicular to the nematic director.

For each sample, a series of intensity autocorrelation functions g(t,q) were obtained at different q values (figure 1). A stretched exponential function was used to fit g(t,q): $g(t,q) = 1 + A[\exp(-\Gamma t)^{\beta}]^2$. For simple diffusion, the relaxation rate Γ yields the diffusion coefficient D via: $\Gamma = Dq^2$.

[1] B. J. Lemaire et al., *Eur. Phys. J. E* **13**, 291-308 (2004)

The dispersion relation obtained for the isotropic, 2 vol% sample at 228K is shown in figure 2a (grey triangles). The relaxation rate increases linearly with q^2, indicating that density inhomogeneities relax via simple diffusion. The diffusion coefficient, D_0, is extracted from the data: D_0=6.4 10^{-16} m^2s^{-1}.

Two other isotropic samples with volume fractions 0.5% and 3.5% have been measured as well (data not shown). The dispersion relation of the dilute sample is very similar to the one for the 2% sample, indicating that D_0 is indeed the diffusion coefficient of isolated, non-interacting particles. However, for the sample at 3.5%, the relaxation rate ceases to be linear in q^2, indicating the onset of interparticle interactions.

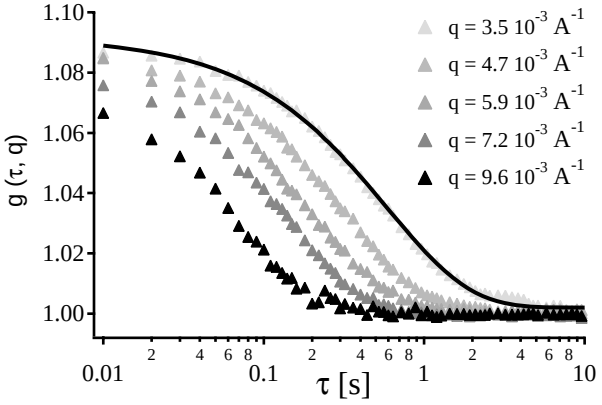

Figure 1: Raw correlation functions (symbols) for 2 vol% isotropic suspension of goethite. The solid black line is a fit by a stretched exponential function with β=0.85.

The dispersion relation obtained for the nematic, 6.7 vol% sample at 228K is shown in figure 2a (black triangles). The relaxation rate is linear in q^2 up to some scattering vector that approximately corresponds to the maximum of the static structure factor S(q). At higher q, the relaxation becomes faster. A linear fit of the Γ values at small q, gives a diffusion coefficient D=1.8 10^{-16} m^2s^{-1}. It thus seems that the repulsive interactions between particles slow down the long-wavelength relaxation.

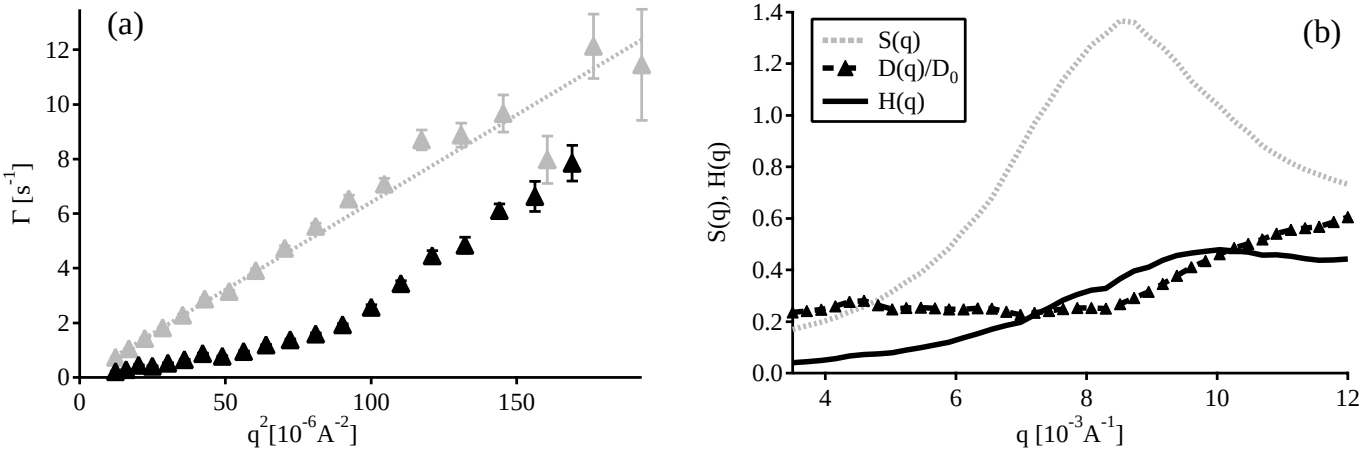

Figure 2: (a) Dispersion relations for a 2% isotropic goethite suspension (grey triangles), and a 6.7% nematic suspension (black triangles). The broken grey line is a fit constrained to go through the origin, that gives D_0 = 6.4 10^{-16} m^2s^{-1}. (b) Normalized diffusion coefficient (grey triangles), static structure factor S(q) (grey line) and hydrodynamic function H(q) (black line) of nematic phase, with q perpendicular to the nematic director.

For a concentrated colloidal suspension with strong interactions, we expect the normalized diffusion coefficient to be equal to the inverse of the structure factor. In the presence of hydrodynamic interactions though, the relation is modified to: $D(q)/D_0 = H(q)/S(q)$, where H(q) is called the hydrodynamic function and gives the indirect interactions between particles. As D(q), D_0 and S(q) have been determined experimentally, we have calculated H(q) for this nematic phase (figure 2b).

In conclusion, we have experimentally determined the hydrodynamic function for a nematic suspension of goethite. Indirect interactions are found to significantly affect particle diffusion, in the q-range containing the peak of the static structure factor. To our knowledge, this function has never been determined for nematic systems before. It is very similar though, to the one measured by XPCS for charge-stabilized spherical colloidal particles[2].

[2] A. Robert et al., *Eur. Phys. J. E* **25**, 77-81 (2008)