| ESRF | **Experiment title:**<br>**Slow dynamics of a colloidal lamellar phase** | **Experiment number**:<br>SC-2690 |
|---|---|---|
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## Report:

The purpose of this experiment was to study the low-frequency relaxation of a lamellar phase consisting of thin platelets, as a function of the particle concentration.

### Experimental

We used suspensions of phosphatoantimonate ($H_3Sb_3P_2O_{14}$) plate-like particles [1], with a typical thickness of 1 nm and at least 300 nm wide. The particles were dispersed in a 50:50 (wt. %) glycerol/water mixture at a volume concentration $\phi$ between 0.5 and 5 %. The samples were prepared by aspiration in round glass capillaries, 1 mm wide and with a wall thickness of 10 µm. By scanning the capillaries in the beam, it is easy to find aligned domains (usually, with the director normal to the capillary walls). The experiments took place at a temperature T = 253 K.

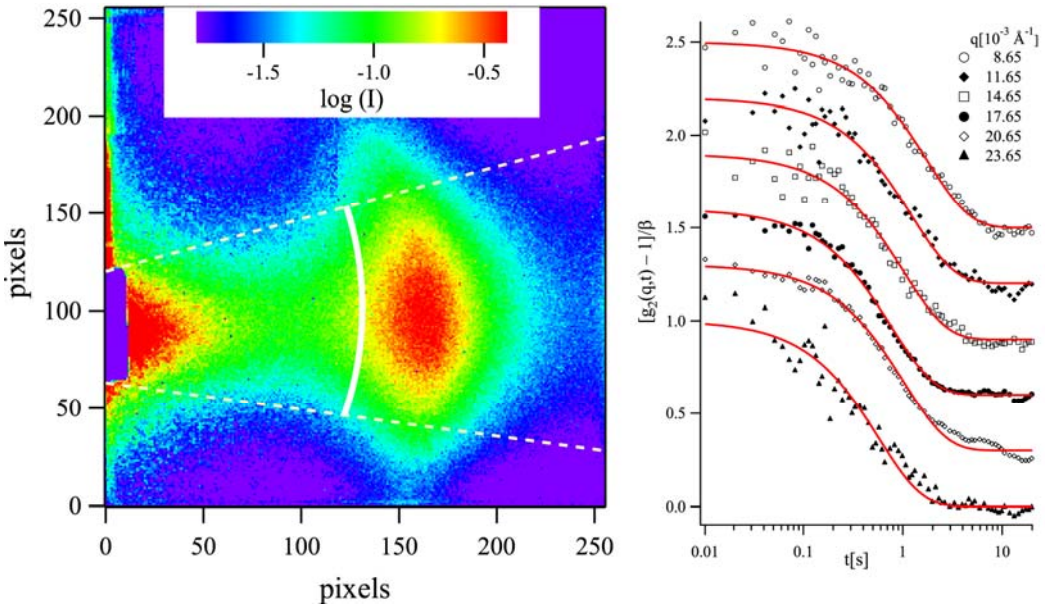


Figure 1: (left) SAXS scattering pattern for the sample with $\phi = 2.1$ vol. %; the lamellar director is roughly horizontal. The averaging was done within the range delimited by the two dashed lines, on circular arcs corresponding to a given scattering vector $q$. One such arc is shown as solid line. (right) Correlation functions for the same sample, at different $q$ values. Solid lines are mono-exponential fits.

The small-angle X-ray scattering (SAXS) and XPCS measurements were performed at the TROIKA beam line ID10A of the ESRF with an X-ray energy of 8 keV ($\lambda = 1.55$ Å) selected by a single-bounce Si(111) monochromator, in the uniform filling mode of the storage ring. A (partially) coherent beam is obtained by inserting a 10 µm pinhole aperture a few centimeters upstream of the sample. We used a 2D Maxipix detector consisting of 256 × 256 square pixels (55 µm in size) and with acquisition rates of up to 1 kHz. For

XPCS, a few thousand images were recorded and the intensity autocorrelation functions $g_2(q,t)$ were calculated by ensemble averaging [2] over pixels with the same absolute value of the scattering vector (see Figure 1, left). Some autocorrelation functions are shown in Figure 1, right for the sample with $\phi = 2.1$ vol. %. The SAXS pattern is obtained as the average intensity over all the images in each run.

## Results

The SAXS data yielded the static structure factor $S(q)$ along the lamellar director (perpendicular to the platelets). Over the position of the first Bragg peak, $S(q)$ is well described by a simple phonon model (point-like particles in one dimension with purely harmonic interaction). The model gives the repeat distance $d$ and the fluctuation amplitude $\sigma$ (related to the spring stiffness). It is noteworthy that for all samples studied, the ratio $\sigma/d$ is close to 0.2.

The correlation functions $g_2(q,t)$ are well described by a monoexponential decay, with a relaxation rate $\Omega(q)$. We define the collective diffusion coefficient $D(q) = \Omega(q)/q^2$ as well as the hydrodynamic function [3] $H(q) = S(q)\cdot D(q)/D_0$, where $D_0$ is the (self)-diffusion coefficient of an isolated particle (Figure 2, left). The high-$q$ value of $D(q)$ is denoted by $D_\infty$ and displayed in Figure 2, right. One can see that $H(q)$ is almost flat at low $\phi$ values but develops a peak similar to that in $S(q)\cdot$as the concentration increases. Over the same $\phi$ range, $D_\infty$ decreases sharply and then follows a slower downward trend.

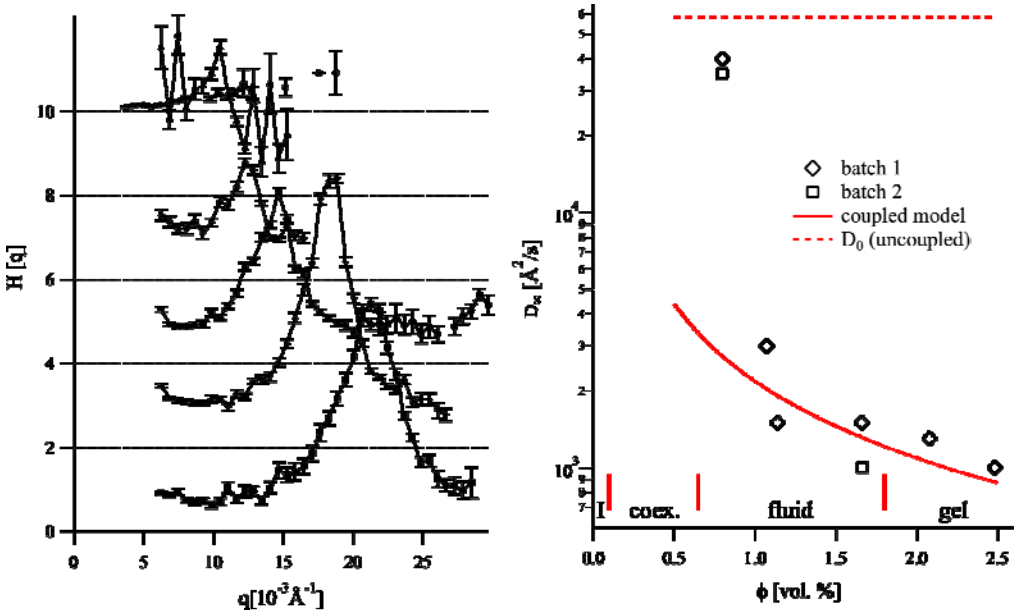


Figure 2: (Left) Hydrodynamic function $H(q)$ for various concentrations $\phi$: 0.8; 1.1, 1.4, 1.7, 2.1 and 2.5 vol % from top to bottom, shifted vertically in steps of 2. (Right) High-$q$ value of the diffusion constant, $D_\infty$, as a function of $\phi$. The phase diagram is also indicated: the (fluid or gel-like) lamellar phase at high concentration and an isotropic (I) phase at very low concentration.

## Interpretation

These features can be explained semi-quantitatively by combining two very simplified dissipation models: In the first one, the particles are uncoupled hydrodynamically, leading to a flat $H(q)$ and a fairly high value of $D_\infty$. In the other extreme, corresponding to strong coupling, the dissipation takes place almost entirely in the squeezing flow between neighbouring particles. In this case, it can be shown that $H(q) \sim S(q)$ and that $D_\infty$ is much lower. The predictions for $D_\infty$ are in fairly good agreement with the experimental data (dashed line for the uncoupled model and solid line for the coupled model in Figure 2, right). Work is in progress on describing the transition between these two regimes [4].

## References


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