



Experiment title:
Dynamic study of fluctuations in lipid multilayer stacks

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Report:

The experiments were performed using an X-ray energy of 13 keV selected by a Si (111) single-bounce monochromator. Highly aligned stacks of several hundreds phospholipid bilayers on silicon substrates have been investigated with a coherent beam. Specular and non-specular scans in and out of the plane of incidence have been performed to characterize the stack. In the range of lateral momentum transfer q_x which was reached by rocking and detector scans in the plane of incidence, a pronounced speckle pattern was observed (Fig.1), indicating that the scattering signal at small $q_x \leq 0.001 \text{ \AA}^{-1}$ is dominated by static defect scattering, probably originating in the domain structure of the lipid film, rather than by thermal diffuse scattering. By using coherent scattering we can thus determine whether static defect scattering or scattering from thermal fluctuation prevails at a given wave vector, which is important also for the interpretation of the incoherent scattering.

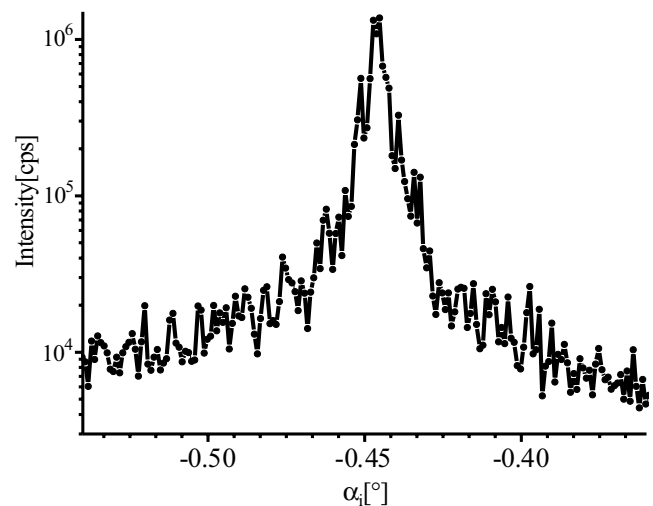


Fig. 1 : Rocking scan through the first Bragg peak (at constant $2\theta = 0.896^\circ$) of a DMPC sample (L_α phase) in excess water. The speckle size and intensity distribution contains information on the domain structure of the film.

XPCS measurements yielded no detectable result for DMPC samples or for the synthetic ionic surfactant DDABr. We were however able to determine the relaxation dynamics for bulk samples of a nonionic mixture, as detailed below.

We measured the dispersion relation $\Omega(q_{\perp})$ in the L_{α} (fluid lamellar) phase of the lyotropic system $C_{12}EO_5$ /hexanol/ H_2O , where $C_{12}EO_5$ stands for the widely studied nonionic surfactant penta-ethylene glycol mono-n-dodecyl-ether. The addition of hexanol as a cosurfactant results in a softening of the bilayers, leading to a bending constant $\kappa \sim k_B T$ [1].

The samples were prepared in 200 mm thick glass capillaries and oriented by thermal cycling between the lamellar and the isotropic phases. Very good homeotropic anchoring is obtained, with the exception of characteristic comb-like structures at the edges and of isolated defects. Measurements were performed at room temperature.

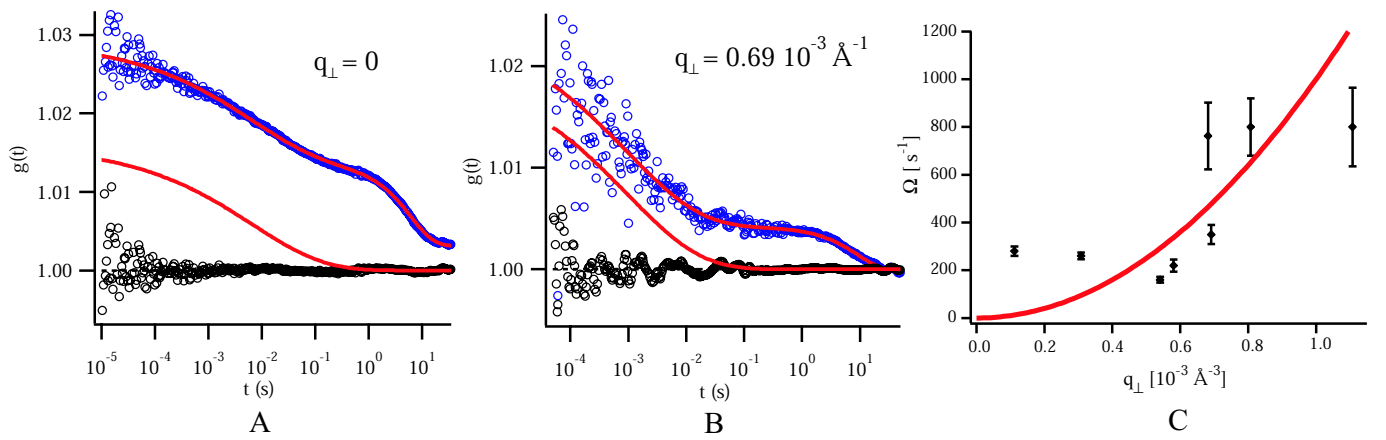


Fig. 2 : A) Correlation function on the first Bragg peak ($q_{\perp} = 0$). B) Correlation function for $q_{\perp} = 0.69 \cdot 10^{-3} \text{ \AA}^{-1}$. C) Preliminary dispersion relation for the undulation mode, for $\kappa = 0.6 k_B T$ [3] and $\eta_3 \simeq 40 \eta_{H_2O}$

The beam was defined by an $8 \mu\text{m}$ pinhole followed by a guard slit for removal of parasitic scattering. The scattered signal was detected by a fast avalanche photodiode (APD) and the output signal was processed online by a FLEX autocorrelator. The measurements were performed in $2 \times 1/3$ filling mode, and the time structure of the ring current is clearly visible in the range $10^{-6} - 10^{-4}$ s. It is extremely important that new measurements be performed in the uniform filling mode, allowing a better fit of the data over a larger time range.

The correlation function $g(t)$ was obtained at each q_{\perp} value by acquiring for 1800 s. After normalization by the autocorrelation of the monitor signal and removal of an oscillatory component due to the mechanical noise of the setup, $g(t)$ was fitted with the sum of a stretched exponential (stretching exponent $\beta \sim 0.35$) representing the relaxation of the undulation mode and a very slow exponential (decay time $\tau \sim 10$ s) of unknown origin: $g(t) = 1 + [a_1 \exp[-(\Omega t)^{\beta}] + a_2 \exp(-t/\tau) + a_3]^2$

The dispersion relation $\Omega(\mathbf{q})$ for fluctuations in the lamellar phase is well known [2]. In the limit of the undulation mode, $\mathbf{q} = (\mathbf{q}_{\perp}, q_z = q_{\text{Bragg}})$, it reduces to $\Omega(q_{\perp}) = \frac{\kappa/d}{\eta_3} q_{\perp}^2$, with κ the bending stiffness, d the lattice spacing and η_3 the layer sliding viscosity.

[1] E. Freyssingeas *et al.*, *Langmuir* **12**, 6028-6035 (1996).

[2] G. Sigaud *et al.*, *J. Phys. II (France)* **3**, 1343-1355 (1993).