Fluctuations in Smectic Membranes

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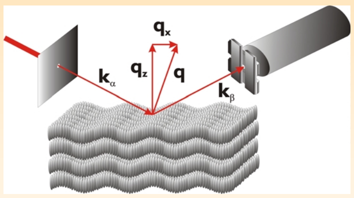
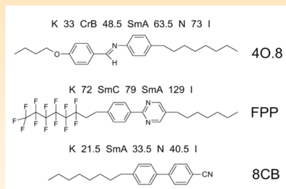
Smectic membranes





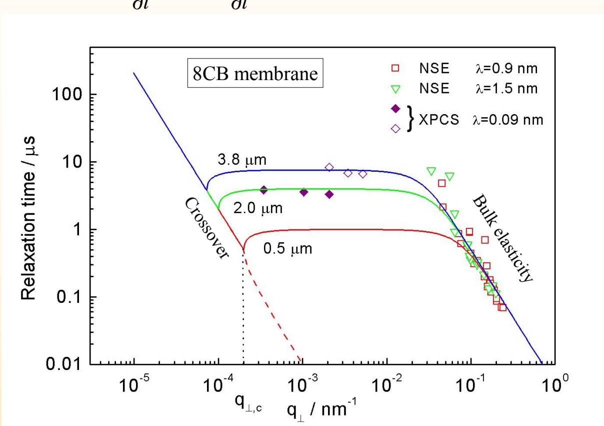
Smectic membranes consist of stacks of liquid layers. Thermal fluctuations disrupt with increasing size of the system the long-range order of the layers (Landau-Peierls instability).

Measurements were done using both x-ray photon correlation spectroscopy (XPCS) at beamline ID10A of ESRF and neutron spin echo (NSE) scattering at beamline IN15 of ILL.

Theoretical outlook

Equation of motion for layer fluctuations u(r,t):

$$\rho \frac{\partial^2 u(r,t)}{\partial t^2} = \eta_3 \frac{\partial}{\partial t} \nabla_\perp^2 u(r,t) + (B\nabla_z^2 - K\Delta_\perp^2) u(r,t)$$



OSCILLATIONS → ← EXPONENTIAL DECAY

High compressibility limit ($B \to \infty$) gives conformal fluctuations

$$\tau_{s,f} \approx \frac{2\rho_0}{\eta_3 q_\perp^2}\left(1 \mp \sqrt{1 - \frac{4\rho_0}{\eta_3^2 q_\perp^4}\left(K q_\perp^4 + \frac{2\gamma}{L} q_\perp^2\right)}\right)^{-1}$$

Crossover wave vector:

$$q_{\perp,c} \sim \frac{1}{\sqrt{L}}$$

$$G(q_\perp,t) = \left\langle u(q_\perp,t) u^*(q_\perp,0)\right\rangle = \frac{k_B T \tau_s \tau_f}{L\rho_0(\tau_s - \tau_f)}\left[\tau_s \exp\left(-\frac{t}{\tau_s}\right) - \tau_f \exp\left(-\frac{t}{\tau_f}\right)\right]$$

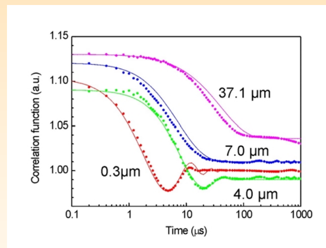
In oscillatory damping mode $\tau_s = \tau_f^*$ (complex relaxation times)

In exponential decay mode $\tau_s >> \tau_f$ fast branch can be neglected:

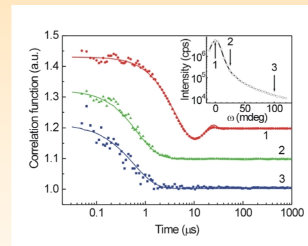
In surface dominated regime (no dependence on $q_\perp$)

In bulk elasticity dominated regime (no dependence on L)

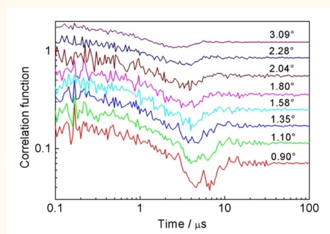
Relaxation in smectic membranes



Thickness dependence for 4O.8 membranes (specular)



Specular and off-specular measurements (see inset) for a 2.83 μm thick FPP membrane

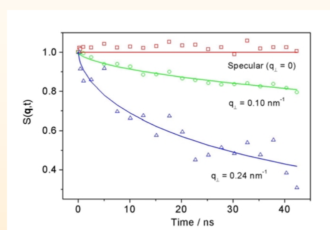


Footprint dependence of relaxation time (29 nm 4O.8 membrane, specular)

The longest detectable wavelength of the fluctuations defines the intensity correlation function. It is related to the size Λ of the coherent scattering volume:

$$I(t) \to \int_{2\pi/\Lambda}^{2\pi/a} dq_\perp e^{iq_\perp r_\perp} G(q_\perp,t)$$

Along the specular ridge the footprint decreases with increasing incident angle.



NSE measurements of a 8CB membrane; results are plotted in the left figure

In NSE due to the low resolution in $q_\perp$ a superposition of exponentials with different relaxation times is found. The correlation functions can be fitted by a stretched exponential:

$$S(q_\perp,t) = \exp\left\{-\left(\frac{t}{\tau}\right)^{0.59}\right\}$$

Conclusions

Using XPCS on the specular Bragg position, for thin membranes oscillatory relaxation behavior is found and for thicker membranes only a single exponential relaxation, in agreement with theoretical predictions.

By changing the scattering geometry to off-specular situations in XPCS experiments one can probe fluctuations at different wave vectors. This results in a crossover between oscillatory and exponential relaxation in dependence of $q_\perp$.

In addition to the surface-tension dominated relaxation probed by XPCS, NSE reveals a new regime at small length-scale fluctuations, determined by liquid-crystalline bulk elasticity.

The transverse coherence length of the beam not only changes the contrast of the correlation function, but also its time dependence. Assuming that the long-wavelength cut-off is defined by the projection of the coherence length on the surface of the film, this behavior can be understood from the theoretical model.

References

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Theory: A.Shalaginov and D.E. Sullivan, Phys.Rev. E **62**, 699 (2000).