ESRF	**Experiment title:** Fluctuations and surface crystallisation in freely suspended smectic films as observed by coherent x-ray scattering.	**Experiment number**: SC-623
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Names and affiliations of applicants (* indicates experimentalists):

Wim H. de Jeu*, Andrea Fera*, Boris I Ovstrovskii*, Igor Dolbnya*‡

FOM-Institute for Atomic and Molecular Physics, Kruislaan 407, 1098 SJ Amsterdam, The Netherlands

‡ DUBBLE CRG, ESRF , BP 220 38043 Grenoble Cedex, France

Report:

The smectic-A phase (SmA) is at its lower marginal dimensionality and characterised by strong thermal fluctuations. As a consequence the positional ordering of the liquid layers is not truly long-range: the mean-square displacement of the layers $u^2(\mathbf{r})$ diverges with the sample size L. Upon cooling such a SmA phase, a 3D crystalline B (CrB) phase can occur, which has an additional in-plane hexagonal structure. Smectic liquid crystals can be suspended over an opening in a solid frame. The resulting free standing films have a high degree of uniformity and a controlled thickness ranging from two to over hundreds of layers. In earlier x-ray reflectivity work we demonstrated that in a SmA film the presence of the free surfaces leads to a strong profile of the fluctuations [1]. This behaviour can be described within the framework the Landau-De Gennes free energy with an additional surface term. When freely suspended smectic films of N-(4-n-butoxybenzilidene)-4-n-octylaniline (4O.8) where cooled from the SmA into the CrB phase still a considerable fluctuation amplitude was found [1b]. This behaviour has been attributed to the small value of C_{44}, the elastic constant for shear of the layers, thus allowing fluctuations similar to SmA films, though not divergent with L.

Recently the dynamical properties of the Landau-De Gennes hamiltonian in the SmA [2,3] and in the CrB phase [3] have been investigated theoretically. In the SmA phase, the relaxation time of the lowest order mode should vary as $\tau_1=\eta_3 L/(2\gamma)$, where η_3 is the shear viscosity of the layers and γ is the surface tension. In fact, a complex spectrum of relaxation times has been predicted, with the other modes depending on the elastic constants for compression (B) and for bending (K) of the layers. For thin CrB films the effect of including C_{44} in the free energy is to renormalize γ by $(\gamma+LC_{44}/8)$ [3]. These relations can be checked by coherent x-ray photon correlation spectroscopy (XCPS). A first pioneering experiment on the dynamics of smectics has been carried out on thick (5–50 μm) SmA films [4]. As experiments with hard x-rays were thought to be impossible, a soft x-ray beam of 0.3 keV was chosen in order to maximise the coherent flux (proportional to λ^2). Despite serious absorption problems, the predicted linear relation between τ_1 and L was nicely confirmed.

We performed XPCS on freely suspended films of 4O.8 both in the SmA and in the CrB phase at 7.98 keV, set by a Si(111) monochromator. A spatially coherent beam was obtained by positioning a 12 μm pinhole at

45 m from the source. The intensity at the detector position was $2x10^8$ photons/s. The beam had a longitudinal coherence length ξ_l=1.5 μm and a transverse coherence length ξ_t=11.3 μm. The thickness of the samples was measured by specular x-ray reflectivity; from rocking curves the mosaicity was determined to be typically of the order of 2 mdeg. The autocorrelation function, measured at the first Bragg position determined by the smectic periodicity of about 2.84 nm, was calculated on line by an ALV-5000 digital autocorrelator operating in the 2-channel mode (0.4 μs time resolution). The detection system consisted of a Ce(YAP) fast scintillation counter at the detector position, and a NaI counter at the monitor position. We reached a time resolution of approximately 0.12 μs for the YAP detector, but only 0.8 μs for the monitor due to afterpulsing effects. The signal from the sample was normalised to the direct beam, recorded between successive acquisitions. As a check, we measured the time autocorrelation function of a NiC multilayer, which was found to be not distinguishable from the direct beam. The data reported in figs.1 and 2 correspond to a relatively thin 0.27 μm film. In spite of being at the edge of the experimental possibilities, a clear difference is found between the SmA and the CrB phase. In conclusion, we have shown that XPCS experiments on freely suspended smectic films are possible in the μs range and with hard x-rays.

In this first attempt severe experimental problems were encountered. (1) The relatively slow correlator available and the poor statistics for lag times $\leq$ 4 μs. (2) The initial attempts to use faster avalanche photodiodes as detectors was not successful because of electronic problems. (3) It turned out to be impossible to use a YAP detector at the monitor position because of parasitic signals. (4) We were not able to make thicker films, which would have allowed to establish a connection with the existing soft x-rays results.

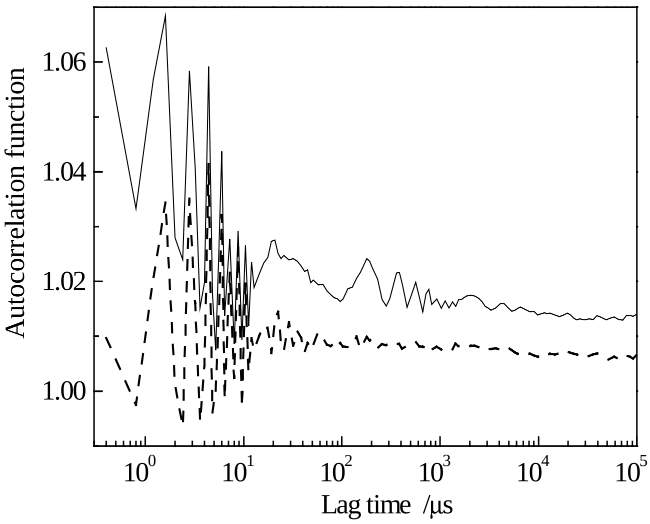

Fig:1: Experimental data for the direct beam (squares) and a CrB freely suspended films (circles)

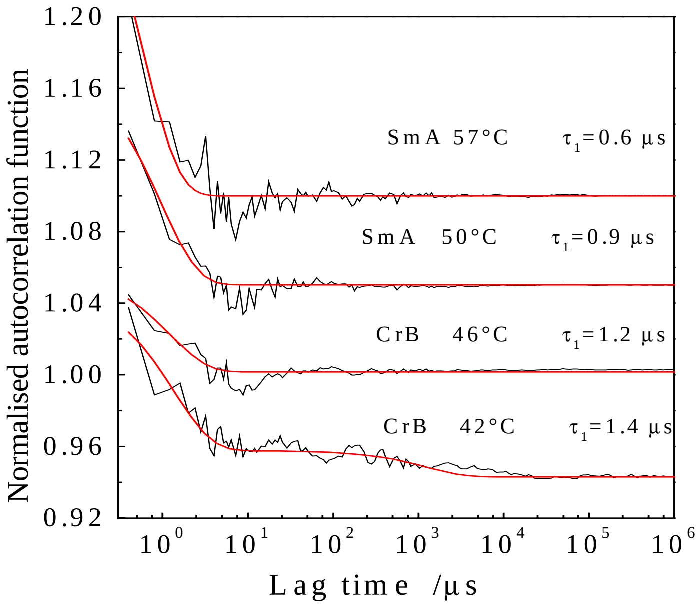

Fig.2: Autocorrelation functions of a freely suspended smectic film of 4O.8 in the SmA and in the CrB phase

References

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