ESRF

Experiment title:
Searching for the loss of conformality in freely suspended smectic films

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SC-191

Beamline:
BM32

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Report:

The fluctuations in freely suspended smectic-A liquid crystalline films have been studied using both specular and off-specular (diffuse) x-ray reflectivity. The former probes the laterally averaged density profile normal to the film (z-direction), the latter the wave vector dependence of the fluctuations. This allows a direct determination of the displacement-displacement correlation function and thus of the surface tension γ, and the elastic constants for compression and bending of the layers, B and K, respectively. Up to now the layers have always been found to fluctuate in unison [1, 2]. The objective of the present experiment was to search for the cross-over from these conformal fluctuations to independent ones. Theoretically this cross-over is expected at an in-plane distance $r_c \approx \sqrt{\gamma L/(2B)}$ [2], L being the film thickness. For wavelengths above r_c the top and bottom of the film should fluctuate independently.

Using a z-axis surface scattering configuration, the diffuse reflectivity has been probed along q_y, normal to the (q_z, q_x) scattering plane. Hence the detector was moved out of this plane keeping the incoming and outgoing angle in the plane constant. This geometry, in combination with the high flux and low background of BM32, allowed us to probe in-plane length scales down to molecular dimensions where $q_y = 1.6$ Å^{-1}, which is about two orders larger than the best previous measurements for such systems. As liquid

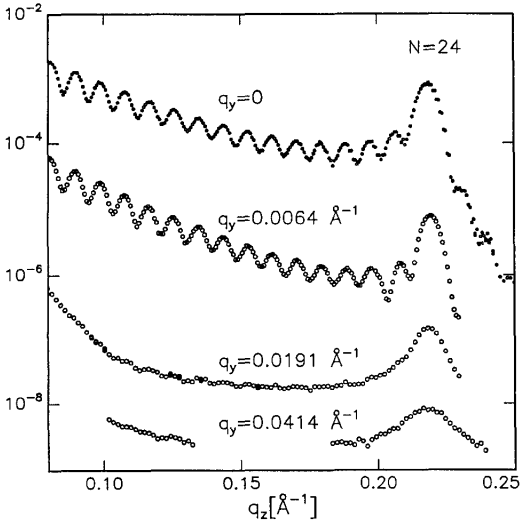

Fig. 1 Longitudinal diffuse scans of a 24 layer film of the compound 7AB at 53.0°C and offsets of q_y as indicated.

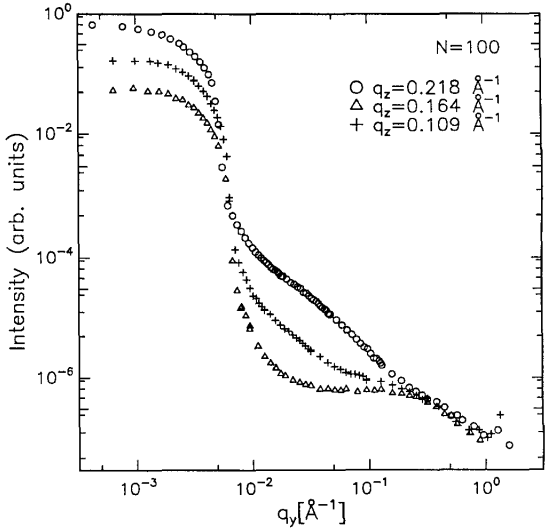

Fig. 2 Transverse diffuse scans at constant q_z (100 layer film of 7AB at 53.0°C). The highest q_z-value corresponds to the first Bragg peak.

crystalline compound *p,p'*-diheptylazoxybenzene (7AB) was used. We worked about 0.5°C below the second-order smectic-nematic phase transition where B could be expected to become small and thus r_c large, and with two film thicknesses (24 and 100 layers).

Longitudinal diffuse scans varying q_z at a constant offset of q_y are shown in figure 1 (background corrected). Note the disappearance of the Kiessig fringes with increasing value of q_y, characteristic of the loss of conformality searched for. At the same time the Bragg peak broadens, which indicates a correlation length smaller than the film thickness. However, the persistence of the Bragg peak implies that correlations between fluctuations of adjacent layers exist down to molecular length scales. More quantitative information can be obtained from the transverse diffuse scans varying q_y at the Bragg peak (q_z=0.218 ~-1), at a subharmonic of the Bragg-peak (q_z=0. 109 ~1), and at an intermediate position (q_z=0.164 Å^{-1}), which are shown in figure 2. Now the loss of conformality is signalled by the changing slope. Further analysis of the data is in progress.

In conclusion, by applying the appropriate scattering geometry to free-standing smectic films close to a phase transition to the nematic phase, we have observed the cross-over from conformal to independent thermal fluctuations.

[1] J.D. Shindler, E.A.L. Mel, A. Shalaginov, and W.H. de Jeu, Phys. Rev. Lett. 74, 722 (1995),
[2] E.A.L. Mel, A. Shalaginov, J.D. Shindler, and W.H. de Jeu, Phys. Rev. E. 54, 536 (1996).