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Report:

We report the result of an experiment where, for the first time, the dynamic properties of a liquid crystal at the Nematic (N) - Smectic A (SmA) transition were probed by X-ray Photon Correlation spectroscopy (XPCS). We have studied the thermotropic liquid crystal 80CB near the N-to-SmA phase transition $(T_{NA} \simeq 337.5K)$, a transition that has previously been characterized in great detail in static, time-averaged X-ray scattering experiments [1]. As the transition is approached from above, small SmA layered clusters are formed in the bulk. In a sample geometry with a free surface, SmA surface layers with essentially infinite lateral extent are formed simultaneously. The viscosity of a nematic liquid crystal is anisotropic and three viscosity coefficients η_{1-3} are defined depending on the relative orientation between the flow velocity \overline{v} , $\nabla \overline{v}$, and the molecular axis \overline{n} [2, 3]. η_3 is diverging at the phase transition while η_{1-2} have simple non-diverging Arrhenius behaviors. In a XPCS experiment [4], a partially coherent beam is required and therefore the beam size was restricted to the transverse coherence length (~ 10 micron). The sample was kept in a closed cell with appropriate temperature control and the amount of 8OCB was chosen such that it in the isotropic phase (T>353K) formed a big flat drop approximately 80 mm wide and 0.5 mm deep. The beam was directed down towards the sample and the incidence angle on the surface was $\theta_i = 0.1^\circ$, well below the critical angle for total external reflection. The scattered intensity was recorded by a scintillation detector mounted on a translation stage at the end of an evacuated flight tube 1227mm down-stream of the sample. The detector signal was processed by an auto-correlator device that calculated the correlation function $g_2(\tau)$ on-line.

The homodyne autocorrelation function $g_2(\tau)$ was found to decay with a characteristic time τ_0 and τ_0^{-1} vs. q_x is shown in figure 1 for T = 338.65 K (solid line). The linear relationship shows that the surface dynamics is governed by over-damped modes. The slope of the line in figure 1 is given by σ/η where σ is the surface tension and η the viscosity. The upper inset in figure 1 shows a sketch of the scattering geometry. The lower inset shows a correlation function and a fit to the expression $g_2(\tau) = Ae^{-\tau/\tau_0} + 1$ (solid line) from which the τ_0 value (40 μs) indicated by the arrow was determined. In figure 2 is plotted the variation of η/σ (found from the slope of the line in figure 1) as a function of reduced temperature t (open circles). The diverging part of the viscosity η_3 can be extracted from the relation $\eta_3/5 = 3\eta - 2\eta_1 - \eta_2$ valid for overdamped modes. η_1/σ and η_2/σ shown in Fig. 2 was taken from Ref. [2]. With T_{NA} as a free parameter, η_3/σ (open triangles) can be well fitted by a power-law with critical exponent $\beta = 0.94$ (solid line). Since the surface tension ($\sigma \approx 28 \text{dyn/cm} [2]$) is almost unaffected by the transition we conclude that the viscosity η_3 of 8OCB shows critical, diverging behavior at the N-to-SmA transition. The result $\beta = 0.94$ is consistent with the theoretical prediction $\beta = 3\nu_{\parallel} - 2\nu_{\perp}$ [5] with the static, critical exponents $\nu_{\parallel} = 0.71, \, \nu_{\perp} = 0.58$ measured by X-ray scattering [1]. However, for unknown reasons there is a striking contrast to the critical exponent $\beta = 0.50$ found by light scattering [2].



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