

## 4. DIFFUSE SCATTERING AND RELATED TOPICS

to all structures mentioned here, there are lyotropic systems in which the smectic-A lamellae seem to break up into cylindrical rods which seem to have the same macroscopic symmetry as some of the discotic phases. On the other hand, it is also much more difficult to prepare a review for the lyotropic systems in the same type of detail as for the thermotropic. The extra complexity associated with the need to control water concentration as well as temperature has made both theoretical and experimental progress more difficult, and, since there has not been very much experimental work on well oriented samples, detailed knowledge of many of these phases is also limited. Aside from the simpler lamellae systems, which seem to have the same symmetry as the thermotropic smectic-A phase, it is not at all clear which of the other phases are three-dimensional crystals and which are true mesomorphic structures. For example, dipalmitoylphosphatidylcholine has an  $L_\beta$  phase that appears for temperatures and (or) water content that is lower than that of the smectic-A  $L_\alpha$  phase (Shipley *et al.*, 1974; Small, 1967; Chapman *et al.*, 1967). The diffraction pattern for this phase contains sharp large-angle reflections that may well correspond to a phase that is like one of the crystalline phases listed in Tables 4.4.1.1 and 4.4.1.2, and Fig. 4.4.1.1. On the other hand, this phase could also be hexatic and we do not have sufficient information to decide. The interested reader is referred to the referenced articles for further detailed information.

## 4.4.6.1. Phases with intermediate molecular tilt: smectic-L, crystalline-M,N

Following the completion of this manuscript, Smith and co-workers (Smith *et al.*, 1988; Sirota *et al.*, 1988) published an X-ray scattering study of the structure of a freely suspended multilayer film of hydrated phosphatidylcholine in which the phase that had been designated  $L_B$  in the literature on lipid phases (Janiak *et al.*, 1979; Luzzati, 1968; Tardieu *et al.*, 1973) was shown to consist of three separate two-dimensional phases in which the positional order in adjacent layers is uncoupled. The three phases are distinguished by the direction of the alkane-chain tilt relative to the nearest neighbours, and in one of these phases the orientation varies continuously with increasing hydration. At the lowest hydration, they observe a phase in which the tilt is towards the second-nearest neighbour; in analogy to the smectic-F phase, they designate this phase  $L_{\beta F}$ . On increasing hydration, they observe a phase in which the tilt direction is intermediate between the nearest- and next-nearest-neighbour directions, and which varies continuously with hydration. This is a new phase that was not previously known and they designate it  $L_{\beta L}$ . On further hydration, they observe a phase in which the molecular tilt is towards a nearest neighbour and this is designated  $L_{\beta I}$ . At maximum hydration, they observe the phase with long-wavelength modulation that was previously designated  $P_\beta$  (Janiak *et al.*, 1979). Selinger & Nelson (1988) have subsequently developed a theory for the phase transitions between phases with varying tilt orientation and have rationalized the existence of phases with intermediate tilt. To be complete, both Fig. 4.4.1.1 and Table 4.4.1.1 should be amended to include this type of hexatic order which is now referred to as the smectic-L. Extension of the previous logic suggests that the crystalline phases with intermediate tilt should be designated M and N, where N has 'herringbone' type of intermolecular order.

## 4.4.6.2. Nematic to smectic-A phase transition

At the time this manuscript was prepared, there was a fundamental discrepancy between theoretical predictions for the details of the critical properties of the second-order nematic to smectic-A phase transition. This has been resolved. Bouwman & de Jeu (1992) reported an X-ray scattering study of the critical properties of octyloxyphenylcyanobenzoyloxybenzoate in which the data were in good agreement with predictions of the three-

dimensional  $xy$  model (Lubensky, 1983; Le Guillou & Zinn-Justin, 1985). The differences between this experiment and others that were discussed previously, and which did not agree with theory, are firstly that this material is much further from the tricritical point that appears to be ubiquitous for most liquid-crystalline materials and, secondly, that they used the Landau-De Gennes theory to argue that the critical-temperature dependence for the  $Q_\perp^4$  term in the differential cross section given in equation (4.4.2.7) is not that of the  $c\xi_\perp^4$  term but rather should vary as  $[(T - T_{NA})/T]^{-\gamma/4}$ , where  $\gamma$  is the exponent that describes the critical-temperature dependence of the smectic order parameter  $|\Psi|^2 \simeq [(T - T_{NA})/T]^{-\gamma}$ . The experimental results are in good agreement with the Monte Carlo simulation of the N- $S_A$  transition that was reported by Dasgupta (1985, 1987).

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#### 4.4. SCATTERING FROM MESOMORPHIC STRUCTURES

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#### 4. DIFFUSE SCATTERING AND RELATED TOPICS

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