New Defect Structures in Liquid Crystals

by

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DECLARATION

I hereby declare that this thesis is composed independently by me at the Raman Research Institute, Bangalore, under the supervision of Prof. N. V. Madhusudana. The subject matter presented in this thesis has not previously formed the basis of the award of any degree, diploma, associateship, fellowship or any other similar title.

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CERTIFICATE

This is to certify that the thesis entitled **New Defect Structures in Liquid Crystals** submitted by P. A. Pramod, for the award of the degree of DOCTOR OF PHILOSOPHY of Jawaharlal Nehru University is his original work. This has not been published or submitted to any other university for any other degree or diploma.

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Preface

Liquid crystals are ordered fluids formed by compounds whose molecules have a large shape anisotropy [1, 2, 3]. In nematics, which are the simplest of liquid crystals, the anisotropic molecules are, on the average, oriented along a common direction. This direction is usually denoted by an apolar unit vector called the director. There is, however, no long range ordering of the centers of mass of the molecules. In the case of smectic liquid crystals, the molecules are condensed into layers. Within the layers there is only a liquid-like order, but along the layer normal the structure is periodic. In smectic-A, the director is along the layer normal. In the case of smectic-C, the molecules are tilted along a common direction within the smectic layers. In presence of chiral (noncentrosymmetric) molecules, the tilt direction in smectic-C varies continuously along the layer normal and is then called a smectic-C^* liquid crystal.

The symmetries of these liquid crystalline structures are intermediate between those of three-dimensional crystals and isotropic fluids. Since they possess a long range orientational order, they can exhibit a large variety of topological defects called disclinations in their director field. Apart from these, smectic liquid crystals can also exhibit dislocalions, as in crystals, due to their layered structure. Since liquid crystals are very soft compared to crystals, effects like chiral interactions play an important role in determining the equilibrium structures formed by them. As a result they can form exotic defect ridden phases when the chiral energy gained by introducing distortions exceeds the elastic energy cost of creating the defects.

In this thesis we describe some experimental and theoretical studies on new defect structures observed in nematic and smectic liquid crystal domains. We also describe the observation of a new liquid crystalline phase, which consists of a network of defects, and give a theoretical model to account for its occurrence.

The topics described in the different chapters are outlined below.

In the first chapter, we give a brief introduction to the various liquid crystalline phases discussed in the thesis. We also describe some of the topological defects commonly seen in various liquid crystals.

In the second chapter, we describe the observation of *spontaneous chiral symmetry breaking* in three-dimensional domains of a smectic-C liquid crystal coexisting with the nematic phase. This new *'chiral'* structure is exhibited by a *binary mixture* consisting of *achiral* molecules. The structure of the smectic domains could be established by studying the optical texture, xray diffraction experiments and optical path difference measurments. The smectic-C domains have a helical structure with a surface disclination line which coils around the domain surface. Also, these domains have highly anisotropic shapes. Experiments performed using an externally applied alternating electric field show that these domains are equilibrium structures. Since the chiral symmetry breaking is spontaneous, both left-handed and right-handed domains form with equal probability. This is the *first* observation of such periodic chiral structures in achiral smectic-C liquid crystals. We could also demonstrate that the application of a chiral bias field in the form of a twist distortion in the director field of the surrounding nematic medium can produce chiral discrimination. The mechanism for this chiral discrimination is easily explained unlike in the case of crystals. The highly anisotropic growth of these domains is also discussed in some detail.

In the third chapter, we present a theoretical analysis of the stability of the chiral domain structure described in the previous chapter. We show that the chiral symmetry breaking is due to a combination of surface anchoring and bulk elastic properties specific to smectic-C liquid crystals. In particular, a cross-coupling between the *twist* and *bend* distortions in the c-vector field, which describes the tilt direction of the molecules within the smectic layers, is responsible for the helical structure exhibited by these domains. This is the *first demonstration* of the effect of such a coupling which is permitted by the symmetry of smectic-C liquid crystal. The sign and magnitude of the corresponding elastic constant are estimated by comparing the theoretical results with the experiments. Experimental observations show that the smectic domains produce a twist deformation in the director field of the surrounding nematic. An estimate of this twist distortion energy shows that it increases rapidly as the domain diameter approaches the thickness of the cell in which the sample is taken. An analysis of the domain shape taking this elastic distortion and the anisotropy in the interfacial tension into account shows that the equilibrium shape is highly anisotropic, as seen experimentally. Also, as the volume of the domain is increased, the domain length increases much more rapidly than the radius, which is also in accord with the experimental observations. We have also simulated the optical texture exhibited by the chiral domains and the results agree reasonably well with the experiments.

In the fourth chapter, we describe the observation of a *new three-dimensionally modulated smectic liquid crystal.* This new phase belongs to the Twist Grain Boundary class of liquid crystalline phases. Twist Grain Boundary (TGB) phases are liquid crystalline analogues of the Abrikosov phase exhibited by type-I1 superconductors. Some of these remarkable structures were theoretically predicted based on an analogy between the two seemingly very different systems [1, 3].

The TGB_A phase, which is the simplest of the TGB phases, consists of a regular twisted arrangement of almost perfect smectic-A blocks separated by grain boundaries consisting of arrays of screw dislocations. TGB phases with smectic-C-like blocks (TGB_C) have also been observed and well studied. The structure of the new phase described in this chapter is far more complicated than the previously observed TGB_A and the TGB_C phases.

We have conducted calorimetric studies to establish the thermodynamic stability of the new structure. Detailed microscopic observations show that the grain boundaries separating the smectic blocks have a two-dimensional height modulation along directions which are orthogonal to the TGB twist axis. We have also performed xray diffraction experiments which show that the local order is indeed like that of smectics with the molecules tilted with respect to the layers. Detailed observations on the response of this structure to an externally applied alternating electric field show that the molecules within each smectic block are arranged in a helical fashion. This is somewhat similar to the molecular arrangement in a smectic-C* liquid crystal. Based on these experiments we have proposed a structure for this liquid crystal. In this, the grain boundaries separating the smectic blocks have a two-dimensional height modulation. Within the blocks, the structure is similar to that of smectic-C*. On going from one smectic block to another the smectic layer normal changes its orientation like in the other TGB phases. Thus the structure is three-dimensionally modulated with twist-axes along three mutually orthogonal directions. Since the grain boundaries have two-dimensional undulations, we call this the Undulating Twist Grain Boundary phase or the $UTGB_{C^*}$ phase, for short.

In the fifth chapter we present a simple model which can account for the occurrence of the various TGB phases. In this the grain boundaries are treated as interfaces with an anisotropic interfacial energy. The smectic blocks are modelled by an ansatz which is chosen based on the experimental observations. The free energy of the smectic blocks is calculated using a Landau type of phenomenological expression. the phase diagrams constructed for different parameters like chirality and anisotropy in the interfacial tension show all the observed TGB phases, including the new $UTGB_{C^*}$ phase.

In the sixth chapter, we present the experimental observations made on a new defect configuration in polymer doped nematic drops.

Nematic drops nucleating from the isotropic phase, or dispersed in another isotropic liquid which is immiscible with the liquid crystal, show a variety of defect configurations. In such drops the director has a preferred orientation at the nematic–isotropic interface, for which the interfacial energy is a minimum. Satisfying the alignment condition on such a closed surface necessarily involves the formation of topological defects (disclinations). The types and configurations of these defects will depend on the elastic properties of the nematic apart from the alignment condition at the boundaries. When the director has a preferrance to lie tangential to the interface, topological cosiderations require the formation of two surface disclinations. In usual drops these disclinations occur diametrically opposite to each other. In the polymer doped nematics, however, the two surface defects lie closer together. When many such drops merge to form large domains, the domains show a periodically distorted structure. Experiments show that the polymer concentration is coupled to the director field of these nematic domains. **A** simple theoretical model which takes into account such a coupling between the gradients in the concentration field and the director distortions does not seem to explain the experimental results. Therefore, other physical mechanisms have to be explored to understand these unusual patterns.

Some of the results in this thesis are available in the following references:

1. P. A. Pramod, Yashodhan Hatwalne and N. V. Madhusudana, "Chiral symmetry breaking in three dimensional smectic-C liquid crystal domains", Phys. Rev. E, 56, R4935, nov. 1997.

2. P. A. Pramod, R. Pratibha and N. V. Madhusudana, "A three dimensionally modulated structure in a chiral smectic-C liquid crystal", Current Science, 73, no. 9, 761, nov. 1997.
   [**cond-mat/9712030**]

3. P. A. Pramod, R. Pratibha and N. V. Madhusudana, "Experimental studies on the Undulating Twist Grain Boundary-C^* phase", In preparation.

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