CHAPTER-1

Introduction

1.1 Liquid Crystal

Conventionally, matter exists in one of the three distinct states of aggregation: the solid state, in which the constituent molecules or atoms have fixed orientation and the centers of mass of the molecules are distributed on a three dimensional periodic lattice that is they possess long range ordering in both position and orientation of the molecules or atoms; the liquid state, constituent molecules or atoms are far apart compared to the solid state and characterized by hindered rotation and translation that is they have no long-range order either in position or in orientation; and the gaseous state, where particles move freely through the entire volume of the container, with almost no constraint to rotation or translation. Besides these three states of matter there exists an important subclass of soft condensed matter known as 'liquid crystal' which was first discovered by the Austrian botanist, Friedrich Reinitzer in 1888 while studying with plant cholesterol **[1-5]**. In liquid crystals the constituent molecules exhibit a certain degree of orientational order and also in addition to orientational order they may have some degree of positional order. The term 'liquid crystal' is due to the fact that these new phases of matter flow like a liquid, and can even form a drop as liquid and at the same time exhibit anisotropic properties (mechanical, optical and structural) of crystalline solid. Because of this intermediate role between fully ordered crystalline solid and completely disordered isotropic liquid, liquid crystal phases are also referred to as mesophases **[5]** **(Figure 1.1)**.

There are fundamentally two different types of liquid crystals: thermotropic liquid crystal, whose mesophase formation is temperature dependent and lyotropic liquid crystals, whose mesophase formation is concentration and solvent dependent. Since no work on lyotropic system has been presented here further discussion will not be made on it. All these intermediate mesomorphic phases differ from each other by orientational and translational order.

Depending on the ordering of the molecules liquid crystals are categorized into two basic classes:

(a) the nematic phase (N), the simplest type of liquid crystal mesophase which arises just below the isotropic phase on cooling which is positionally completely disordered but the molecules have a long-range orientational order;

(b) the smectic phases, which in addition to the orientational order possess a reduced translational order than the crystals and arises on cooling usually form nematic phase. Actually smectic phases forms a two dimensional layered structure and within the layers the molecules are distributed without any or with some degree of translational order (sometimes termed as two-dimensional liquid) **[5]**. Depending on the degree of orientational and translational order there exist four sub-groups of smectic phases. In first two groups the molecules have their long axes on an average normal to the layers and are distinguished by the degree of positional ordering of the constituent molecules. For example in smectic A or in hexatic B phase though the molecules have their long axes nearly normal to the layers but they have very short range positional ordering **[6]** whereas in crystal B or in crystal E phases the constituent molecules have long range positional ordering in 3-dimension **[7]**, of course not like crystals. The other two sub groups have their constituent molecules tilted relative to the layer planes such as in smectic C, smectic I and smectic F phases and the molecule have short range positional ordering **[8,9]** while in crystal G, crystal H, crystal J and crystal K phases the molecules have long range 3-dimensional ordering **[9,10]**. The alphabets were assigned to different phases simply in chronological order in which these phases were discovered. Many books and review articles describing the various phases and properties of liquid crystal are available in the literature **[Appendix A]**. From the current knowledge the sequence of liquid crystalline phases in decreasing ordering or with increasing temperature may be written as follows **[11]**:

Crystal→H →K →E →G →J → SmF → B(cryst)→[B(hex), SmI] →SmC →SmA→N(or Ch) → Isotropic Liquid.

Smectic A, C, B_{hex}, I and F are smectic liquid crystals while B, E, G, H, J and K are termed crystal phases. The latter phases however have properties different from normal crystals. Different types of smectic phase structures have been investigated by X-ray diffraction techniques [11-16].

Again when the constituent molecules of a liquid crystal are chiral (if the mirror image of an object cannot be made to coincide with the original object then it is called chiral), liquid crystal mesophases designated as cholesteric (N*), chiral smectic A (SmA*), chiral smectic B (SmB*), chiral smectic C (SmC*), etc. In this thesis chiral smectic phases will play the lead role.

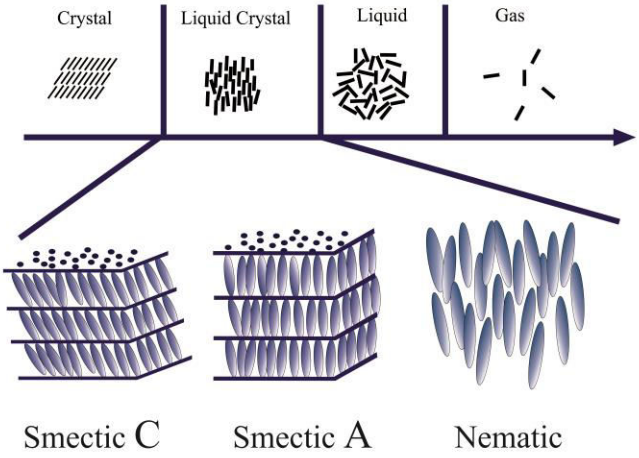


Figure 1.1: Liquid crystal phase in between crystal and liquid phase. Drawn after I. Dierking **[Appendix A]**.

1.2 Ferroelectric Liquid Crystal (The SmC* phase)

A material is said to be a ferroelectric if it possesses a spontaneous polarization in absence of external field and polarization is switched between +ve and –ve state with the application of an electric field. In general, some special types of solid materials which have non-centro symmetric space group show ferroelectricity [17,18]. But, way back in 1974, using elegant symmetry considerations Robert Meyer showed that ferroelectric properties could be exhibited by liquid crystals of low symmetry such as in the chiral smectic C (SmC*) phase [19, 20]. Molecular structures and phase sequences of first FLC and AFLC compounds are shown in **Table 1.1**.

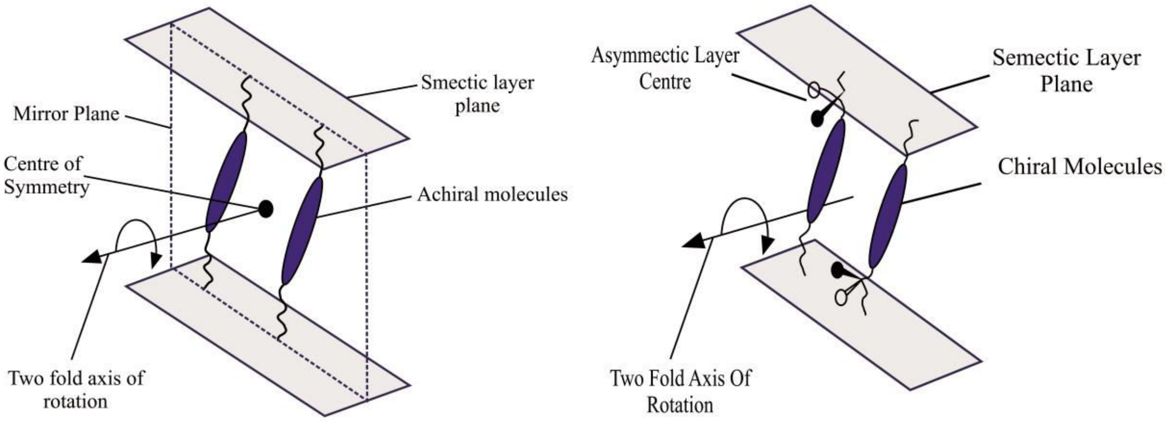


Figure 1.2: (a) C_{2h} symmetry of SmC phase (b) C_2 symmetry of SmC* phase. Drawn after M. Hird [21].

From the **Figure 1.2(a)**, it is clear that an achiral smectic C (SmC) phase possesses C_{2h} symmetry operation, that is, SmC phase has three elements of symmetry: (i) a two-fold axis of rotation normal to the tilt direction in the plane of the layer (ii) a mirror plane normal to the two-fold axis and the smectic layer plane and (iii) a centre of symmetry. But when the constituting molecules of the SmC phase are chiral, the symmetry operation is reduced to two-fold axis of rotation [21]. Hence, SmC* phase described by C_2 symmetry operation as shown in the **Figure 1.2(b)**. Due to this reduced symmetry, SmC* phase possesses a spontaneous polarization (P_S) in absence of external electric field. On application of an external electric field molecules switches around an imaginary cone such that the sign of the P_S gets reversed **(Figure 1.3)** and would remain in the same condition even if the field is removed. The sign of the P_S can again be reversed to the initial state when an electric field of opposite direction is applied. Thus, the SmC* phase exhibits ferroelectricity. Switching of the molecule from one orientation to another in presence of electric field simultaneously changes the optical effect of the materials and mainly this electro-optical effect is employed in liquid crystal based display applications.

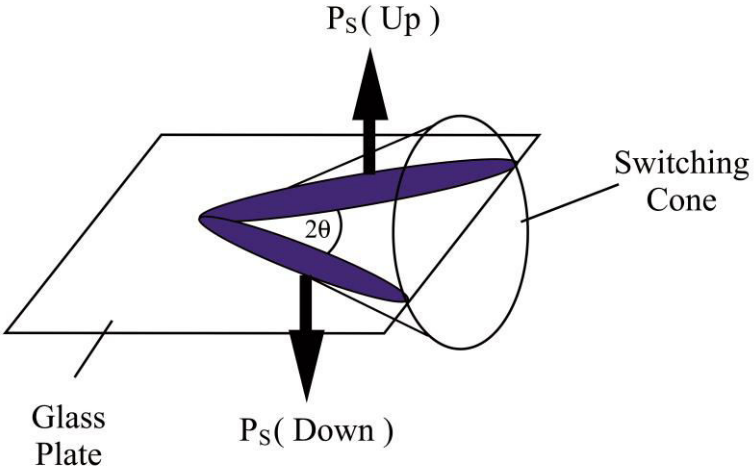


Figure 1.3: Bistability of SmC* phase. Drawn after M. Hird **[21]**.

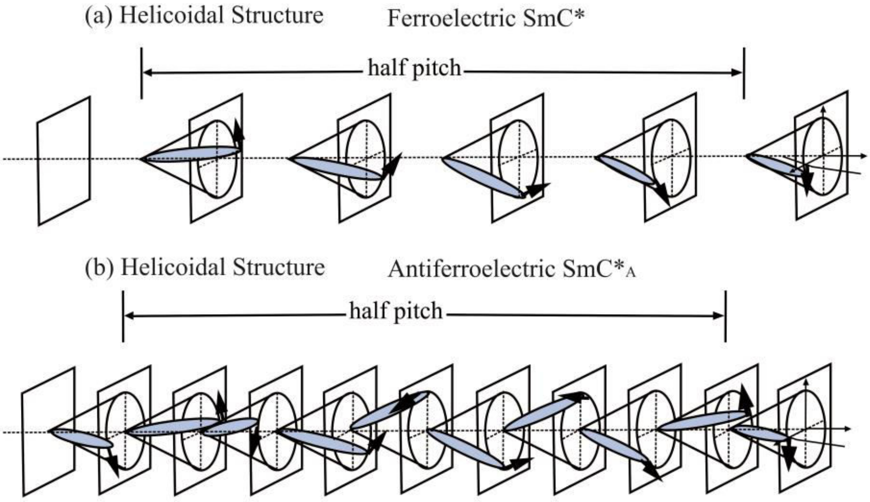


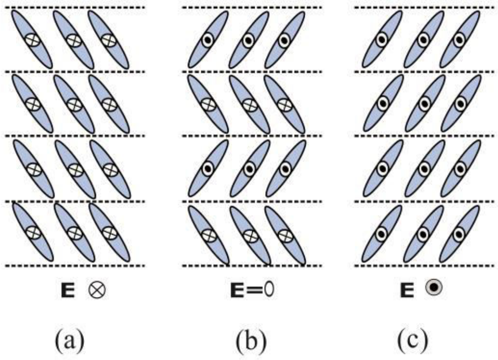
Figure 1.4: Helicoidal structure of (a) SmC* phase and (b) SmC_A* phase. Drawn after H. Takezoe **[30]**.

Moreover because of the chirality of the molecules the bulk SmC* phase constitute a helical structure in which the molecular tilt-angle is constant throughout the sample, but the phase-angle changes gradually from layer to layer which in turn changes the direction of the P_S in the same way (**Figure 1.4(a)**), and hence P_S cancels to zero in the bulk SmC* phase. However, SmC* phase can be used in ferroelectric applications if one can unwound the helix. Unwinding the helix by surface interaction, Clark and Lagerwall first reported the ferroelectric display device which is known as surface stabilized ferroelectric liquid crystal (SSFLC) display device [22].

1.3 Antiferroelectric Liquid Crystal (The SmC$_A$* phase)

Antiferroelectric liquid crystal (SmC$_A$*) is a subclass of previously described ferroelectric liquid crystal (SmC*). In SmC$_A$* phase, director tilt changes sign from layer to layer that is it exhibits an anticlinic structure (**Figure 1.4(b)**), unlike SmC* phase which exhibit synclinic structure. Now as the phase is chiral spontaneous polarization also changes sign as one move from one layer to another. Like the ferroelectric SmC* phase bulk antiferroelectric SmC$_A$* phase also form helical structure (**Figure 1.4(b)**), but in this case antiferroelectric behavior is independent of the helix and is inherent to the phase itself, whereas ferroelectricity is only a consequence of the unwinding of the helix in the SmC* phase.

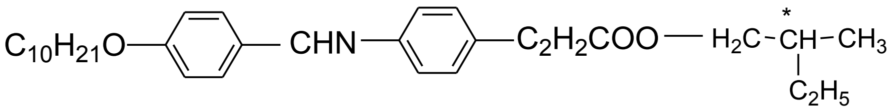
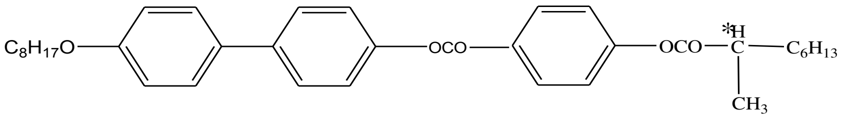
By applying a minimum electric field known as threshold field, an anticlinic antiferroelectric state can be changed to synclinic ferroelectric state. So switching of the antiferroelectric SmC$_A$* phase has three stable states: (a) the anticlinic antiferroelectric state (b) the synclinic ferroelectric state with +ve spontaneous polarization and (c) the synclinic ferroelectric state with -ve spontaneous polarization, as shown in the **Figure 1.5**. As SmC$_A$* phase has two switched states which correspond to opposite polarities of the applied field a global DC-compensation is easily achieved and posses a natural grey scale which are the necessary conditions for high resolution low power high speed liquid crystal display [23].



(a) (b) (c)

Figure 1.5: The local molecular arrangements of (b) the antiferroelectric phase, (a) the positive and (c) the negative electric field-induced ferroelectric phases. Drawn after H. Takezoe [30].

Table 1.1 Molecular structures, phase sequences and transition temperature (°C) of first FLC and AFLC compounds.

Samples	Molecular structure and transition temperatures
FLC: DOBAMBC [19]	 Cr 76 SmC* 95 SmA* 117 I
AFLC: MHPOBC [24]	 Cr 84 SmC_A* 88.4 SmC_γ* 119.2 SmC* 120.9 SmC_α* 122 SmA*148 I

1.4 Para-electric liquid crystal (The SmA* phase) and Electroclinic Effect

The Smectic A* (SmA*) phase is the simplest chiral liquid crystal phase in which the long axis of the molecules (director) is parallel to the layer normal and that is why this phase is termed as orthogonal SmA* phase. In general without an external perturbation chiral SmA* phase cannot be distinguished from its achiral version SmA phase. But when an electric field E is applied parallel to the layers of chiral SmA* phase, it induces a tilt of the molecular axes in a plane perpendicular to the field direction. The induction of tilt in SmA* phase arising from the coupling of transverse dipole moment of chiral molecule to an applied electric field is known as electroclinic effect (ECE) and was first observed by Garoff and Meyer **[25,26]**. At low electric field induced tilt show linear dependence and at a given magnitude of electric field induced tilt is higher near SmC*-SmA* transition and decreases as the temperature increases. Also the liquid crystal compounds which have high values of spontaneous polarization in SmC* phase exhibit large electroclinic effect due to the induced polarization in the SmA* phase.

1.5 Applications of Liquid Crystals

Optical birefringence, dielectric anisotropy, elastic constants, viscosity coefficients of liquid crystals are sensitive to weak external stimuli such as electric and magnetic fields, heat energy and acoustical energy etc. Due to these distinctive physical properties liquid crystals are widely used in various scientific and technical applications. For example, a small change in temperature changes the pitch of the helix of a cholesteric liquid crystal which results in a color change. For this cholesteric liquid crystals are used in thermal mapping of human skin for diagnosis of circulatory system diseases or for the detection of tumors etc **[27]** and also in thermal mapping of electronic components of an integrated circuit **[28]**. Cholesteric liquid crystals have also been used to measure surface temperatures in aerodynamic designs. Liquid crystals also make good stationary phases in gas-liquid chromatography (GLC), for this they used as solvent for separation of two geometric isomers such as m- and p-xylene. As liquid crystalline solutions with non-mesogenic guest molecules are easily oriented in electric or magnetic field that is why they are also used as solvent for organic molecules in nuclear magnetic resonance (NMR) measurements and infrared (IR) or ultraviolet (UV) spectroscopy to perform polarization studies.

Though liquid crystals are used in various areas of science and technology but due to relative ease and efficiency of excitation with an external electric or magnetic field as compared to other means of excitation (as discussed earlier) most of the present research is centered on the application of electro-optic effects especially on display applications. Liquid crystal electro-optic effects are related to the switching of the molecules with respect to applied electric field **[29,30]** and not require any emission of light they just modify the passage of light. This type of electro-optic switching allow direct reading of symbols, letters, numbers and can display more information in less space and device configuration is known as liquid crystal display (LCD). LCDs are most widely used displays as they exhibit low input power consumption per unit area, wide-angle viewing, fast switching time, large area display by small volume, design flexibility, high resolution, wide temperature range and high contrast ratio compared to other displays. Other than display applications, electro-optical switching is also technologically very useful for devices like spatial light modulators, telecommunications switching, optical recognition and optical computing, micro-color filters, tunable color filters etc **[31-37]**.

1.6 Liquid Crystal Display

Now-a-days displays have become a field of tremendous importance as they provide the best means for interface between man and machine **[38,39]**. Though large numbers of displays are presently available in the market, but among all displays not less than 90% of that market is controlled by the liquid crystal display (LCD) technology **[39]**. Almost in all kinds of optical applications, we use in our daily-life, such as watches, pocket calculators, note books, personal digital assistants, mobile phones, digital cameras, desktop monitors, laptops and TVs, liquid crystal displays are used [40-42].

Though liquid crystals are invented in 1888, it was Vsevolod Fréedericksz who first devised the electrically switched liquid crystal light valve, known as Fréedericksz transition, in 1927 [43]. Many years after that in 1964, first operational liquid crystal display based on dynamic scattering mode (DSM) was invented by G. H. Heilmeier [44,45]. In 1970, twisted nematic-field effect (TN-FE) was introduced by W. Helfrich and M. Schadt and during this time displays were produced for wrist watches using this TN-FE [46]. In 1971, LXD Inc. produced first liquid crystal displays based on TN-effect with low operating voltage and low power consumption compared to poor quality DSM types LCDs. Then T. P. Brody and his team introduced active matrix LCD panel for the first time in 1972 [47] and 11 years after that super twisted nematic (STN) structure for passive matrix addressed LCD panel was invented by H. Amstutz and his co-workers [48]. In 1984, T. Welzen and A. de Vaan, two Philips researchers, developed high speed driving schemes enabling to produce high resolution, high quality, and smooth moving video images on STN-LCDs [49]. To solve the problems related to viewing angle and low quality color production of TN and STN-LCDs, a completely different type of electro-optical effect was introduced, in 1990, known as in plane switching mode (IPS-mode) [50]. There after Hitachi further optimized the IPS technology and manufactured active matrix addressed LCDs which is a milestone for implementing large screen LCDs for flat panel computer monitors and TV screens [51]. In 1996, Samsung developed multi-domain LCD, this multi-domain LCD together with IPS technology remain dominant in LCD design technology and totally surpassed the image quality of cathode-ray-tube-based (CRT) displays [52]. At present IPS is used in the vast majority of smart-phones, tablets, computer monitors and of course in TVs, but research is still continue to produce thin, large sized, high definition (HD) or ultra HD LCDs with maximum viewing angles [53].

1.7 Motivation of the Present Work

Today though LCD technology completely dominates the display market, but there are some other display technologies such as OLED (Organic Light Emitting Diode), plasma etc. based displays which are slowly making their way to being the solid competitors in the display market. To maintain its dominant role in the display industry LCD technology should be improved time to time and for this one requires improved liquid crystal materials together with advanced driving technology. Advance liquid crystalline material had to be developed in order to fulfill the requirements of higher resolution and large size LCDs. Most of the LCDs which are discussed in the earlier section mainly nematic liquid crystal based displays (NLCDs) i.e.; they uses nematic liquid crystal as the display material. Main problem in NLCD is the relatively low switching time (of the order 20 ms to 10 ms) which results in a low contrast ratio of the display device **[47-49]**. Such type of low response also narrows the viewing angle when driven at a high multiplexing ratio which is necessary for high information content display devices. Again it is very difficult to obtain memory states as well as sharp threshold voltages for electro-optic effects in these NLCDs **[23]**.

The problems of NLCDs are removed to a large extent if we use ferroelectric liquid crystal (FLC) or antiferroelectric liquid crystal (AFLC) as display materials **[54-58]**. Current research shows that FLCs have potential to become the best choice for displays as they exhibit very fast switching (microsecond order switching time) and switching occurs in an in-plane manner, which provides for excellent angles of view that were not possible for nematic displays **[59-62]**. Because of inherent bistability of a ferroelectric device, almost no power is required to maintain the image of the display **[23]**. Like FLCs, AFLCs are also very promising for high-resolution displays due to their tri-state switching behavior, steep threshold, microsecond response, easy direct current compensation, hemispherical viewing angle and inherent grey scale capability **[55,58,62-65]**.

Though FLC and AFLC materials are very promising for high resolution full color displays, but still FLC or AFLC displays are not commercialized much. Because, to meet consumer expectations that is in order to achieve desired properties of a display device we have to optimize all the requirements like the temperature range, rise time, birefringence, polarization, tilt angle, length of pitch, viscosity, thermal and UV stability etc of FLCs and AFLCs **[21,66-68]**. But a single FLC or AFLC compound cannot satisfy all such requirements necessary for display applications.

Multi-component liquid crystal mixtures are, therefore, formulated to optimize all the required properties for practical applications. Multi-component mixture consists of two components: (a) achiral host material and (b) chiral dopant. The host mixture normally is a mixture of several achiral compounds with a reasonably wide SmC temperature range starting from room temperature or below, which control the overall temperature range and tilt angle of the final mixture. Dopant is a compound with one or two chiral centers which control the spontaneous polarization, viscosity, switching speed and helical pitch of the final mixture **[69-73]**. It might be noted that every component influences the properties of the resulting mixture, a particular property will depend not only on the type and proportion of chiral dopants, but also on the host material used. However, the dopants are used to optimize the final properties to the desired level more effectively.

So to formulate a ferroelectric mixture with desired properties one has to first prepare a low viscous host mixture with a wide SmC phase at or close to room temperature preferably together with SmA and nematic N phase from ease of alignment point of view and then select a proper chiral dopant having high spontaneous polarization but at the same time which imparted low viscosity to the mixture. In host mixture presence of SmA and nematic N phase are important because chiral manifestations of these phases i.e., SmA* and N* play the lead role for obtaining better alignment of the molecules in bookshelf geometry in display cells **[21,22,74]**. In literature many reports are available on various properties of room temperature ferroelectric and antiferroelectric liquid crystal mixtures **[70-78]** which are commercially are available [**Appendix A**]. But the fact is that in most of these publications, commercial mixtures with unknown compositions were used. Therefore, it is impossible to extract information about how the structures chiral liquid crystal materials influence ferroelectric and antiferroelectric properties in achiral host material. This information is important in order to find the right way to design mixtures with good properties for display devices.

The main objectives of the present dissertation are:

1. *Formulation and characterization of binary as well as multi-component ferroelectric and antiferroelectric liquid crystal mixtures using proper achiral host materials and different chiral dopants.*
2. *To study how the molecular structures of chiral dopants affect the mixture property.*
3. *To charecterize the formulated mixtures to find their suitability for display applications.*

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