ELECTRO-OPTIC RESPONSE OF HOMEOTROPIC, HOLE PATTERNED, NEMATIC LIQUID CRYSTAL CELLS UNDER AC AND DC VOLTAGE

A dissertation submitted
to Kent State University in partial fulfillment of the requirements for the degree of Doctor of Philosophy

by
Souptik Mukherjee
August 2016

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Dissertation written by
Souptik Mukherjee
B.E, National Institute of Technology Karnataka, India, 2003
M.S., University of Texas at Arlington, 2008

Approved by

___________________________________, Chair, Doctoral Dissertation Committee
Dr. Hiroshi Yokoyama

___________________________________, Members, Doctoral Dissertation Committee
Dr. Bahman Taheri (Advisor)

_________________________________
Dr. Ling-Chy Chien

_________________________________
Dr. Elizabeth Mann

_________________________________
Dr. John West

_________________________________
Dr. Roger Gregory

Accepted by

___________________________________, Chair, Doctoral Dissertation Committee
Dr. Hiroshi Yokoyama

___________________________________, Dean, College of Arts and Sciences
Dr. James L. Blank
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**TABLE**

Table 1. Critical voltage required to re-orient the LC molecules outside the hole etched region and the voltage corresponding to the completion of the re-orientation in the hole etched region for different hole diameters ranging from 1mm to 4mm.
ACKNOWLEDGEMENTS

I would like to acknowledge the support of Dr. Bahman Taheri, Dr. Antonio Munoz and Dr. Hiroshi Yokoyama for his guidance, support and encouragement. This thesis would not have been possible without their support and proper guidance. I would also like to express my gratitude to my beloved family for their understanding and endless love, through the duration of my studies.

Souptik Mukherjee

07/05/2016, Kent, Ohio
CHAPTER 1

Introduction

Liquid crystals are used in a variety of electro-optic applications. One such application is use of liquid crystals for phase front correction which can take the form of liquid crystal lenses or retarders. An interesting aspect of these devices is the need for inhomogenous electric field for their operation. A number of studies have been performed to investigate the performance of liquid crystals in these configurations over the past decade [1-5].

It is well known that an external electric field influences the director orientation of liquid crystals (LC) [6,7]. This alters the refractive index of materials which in turn can alter the phase and intensity of an incident light. These effects have already been reported [8,9] under a homogenous electric field. More recently, the electro-optic response of liquid crystal cells with patterned electrodes has been reported by various groups [10-13]. One simple electrode patterns used is the so-called hole-patterned electrode. The hole-patterned electrode has been studied as a hole patterned capacitor filled with a electro-rheological fluid. The reported size of the apertures in the hole patterned electrode ranges from 300μm to 7mm [14-15]. The hole patterned electrodes create an inhomogeneous electric field which then influences the orientation of LC materials [16]. Other parameters like the thickness of the cell [17], the birefringence of the LC, the polyimide used in the surface alignment and the dielectric anisotropy of the LC materials may influence the electro-optic properties [18-21]. To understand the phenomenon associated with LC cells having patterned electrodes, the properties of LC materials need to be understood and optimized. The electro-optic responses associated with patterned LC cells, reported so far, are primarily associated with AC voltage.
The initial goal of the research was to further this knowledge and development as it applied to liquid crystal lenses. However, as part of the initial experiments, I observed a unique phenomenon. In particular, contrary to the common understanding, I observed that the response of the system was significantly different if the applied field was AC or DC. Given that liquid crystals response is expected to be indifferent to this, the observation provided a unique divergence from the initial goal. As such, the focus of the research was changed to investigation of this phenomenon.

In addition to the work above, a cell with fluorescent dye was fabricated and used for a liquid crystal random lasing experiment. This work was published along with other colleagues. Since that work was beyond the initial effort, I have placed that in the appendix. This dissertation summarizes the results of my experiments in this field. The thesis is divided into three chapters. The first chapter is introduction to the work done by me in this dissertation which has not been reported before. The second chapter deals with background information of liquid crystals and hole patterned LC cells devices which have been previously used in the literature to produce an inhomogenous electric field. Various types of hole patterned liquid crystal hole patterns have been discussed and in this chapter we provide a brief summary of the relevant work. The third chapter deals with the fabrication of hole patterned liquid crystal cells. In particular, I present the geometry I used to create the hold pattern including the alignment layer, cell gap and diameter of the hole pattern. The fourth chapter deals with the results obtained from the experiments performed. In that chapter, I introduce the results from different voltages including the application of AC and DC voltage for cells with different PI thickness and different size of hole diameters. The phase profiles were measured and analyzed in terms of phase retardation.
Chapter 2

BACKGROUND

2.1 Nematic LC in optical systems

2.1.1 Liquid Crystalline State of Matter

Liquid crystal materials consist of organic molecules having characteristic anisotropic shape. They are an intermediate phase of matter and has properties between crystalline (solid) and isotropic (liquid) states. In other words liquid crystal molecules have orientation like a crystal but flow like a liquid. This unique property of liquid crystals make them suitable for a number of applications including liquid crystal displays (LCDs).

Liquid crystal phases can be broadly classified into thermotropic, lyotropic and phototropic LC phases. Thermotropic liquid crystals are those in which the phase transitions are temperature sensitive (fig. 1.1). In case of lyotropic liquid crystals the phase transitions are based on both the temperature and the concentration. Phototropic LC change phase when subjected to light [1-2].

Based on the shape of the molecules, liquid crystals can be classified into different types. If the shape of the liquid crystal is rod like, then it is called calamitic liquid crystals while it is called discotic liquid crystals if the shape of the molecules are disc type. The liquid crystalline phase is also determined by the degree of order of liquid crystal molecules. Two kinds of order are important. One is the orientational order and the other is the positional order. Orientational order shows the extent to which the liquid crystal molecules are aligned along its director. The positional order shows the extent to which the average liquid crystal molecule or group of molecules have
long range translational symmetry. Some liquid crystal phases like smectic liquid crystals have both orientational order as well as positional order. In this dissertation I work with nematic liquid crystal materials. Nematic materials have no positional order but have long range orientational order. Nematic liquid crystals belong to thermotropic liquid crystal materials and are thereby sensitive to temperature. Nematic liquid crystals are also uniaxial materials having one long axis and two mutually perpendicular equivalent axes.

Figure 2.1. Thermotropic liquid crystal phases occur between solid phase and the liquid phase and are sensitive to temperature. Reproduced from [1], p.2.

### 2.1.2 Director

The director of the liquid crystal molecules refer to a direction that the long molecular axis of all molecules, averaged over time, prefer to align even though there is constant fluctuation of the molecules. The liquid crystal director is non-polar, which is a consequence of the mirror symmetry that exists in the molecules in the nematic liquid crystal phase. It is represented by a unit vector \( \mathbf{n} \). Being non-polar, the unit vector \( \mathbf{n} \) is equivalent to \(-\mathbf{n}\) [3].
2.1.3 Anisotropic Physical Properties

The rod like shape of the liquid crystal molecules provide physical properties which are anisotropic in nature. Also because of the uniaxial symmetry of the liquid crystal molecules the measured physical properties parallel to the long axis of the molecule, provided by the director, is different from the two perpendicular directions. The measured physical properties along the two perpendicular directions are equivalent.

2.1.3.1 Dielectric Anisotropy

The response of the liquid crystal molecules to an applied electric field is determined by the dielectric anisotropy of the liquid crystal molecules. When the electric field oscillates parallel to the long axis of the liquid crystal molecules, the dielectric constant is given by $\varepsilon_{\parallel}$. When it vibrates perpendicular to the long axis of the molecules the dielectric constant is given by $\varepsilon_{\perp}$ (fig.1.2). The dielectric properties of a liquid crystal material is given by a tensor as given in eq. 1.1.3.1.1 [3]. The dielectric anisotropy ($\Delta\varepsilon$) is given by eq. 1.1.3.1.2 [3]. When the dielectric anisotropy is positive, i.e when $\varepsilon_{\parallel} > \varepsilon_{\perp}$, the liquid crystal molecules align along the direction of the applied electric field. This happens because in case of positive dielectric anisotropy, the polarizability of the liquid crystal molecules is more along the long axis of the molecules and therefore the molecules align in the direction of the electric field. On the other hand, when the dielectric anisotropy is negative, i.e when $\varepsilon_{\parallel} < \varepsilon_{\perp}$, the molecules align perpendicular to the direction of the electric field. In this case, the polarizability of the liquid crystal molecules will be perpendicular to the direction of the electric field (fig.2.3).
The dielectric anisotropy is given as [3]

$$\Delta \varepsilon = \varepsilon_\parallel - \varepsilon_\perp$$  \hspace{1cm} (2.1.3.1.2)

The free energy density of the liquid crystal is an extremely important parameter. The contribution of the electric field to the free energy density is given by eq. 2.1.3.1.3. In eq. 2.1.3.1.3, $f$ is the free energy density, $D$ is the electric displacement, $E$ is the electric field and $\hat{n}$ is the unit vector representing the director. The free energy density is obtained using the relation between the electric displacement and any arbitrary electric field is given by eq. 2.1.3.1.4. The free energy term in eq. 2.1.3.1.3 shows that for materials with positive dielectric anisotropy, for the free energy value to be minimum, the liquid crystal molecules will tend to align with the direction of the electric field. When the dielectric anisotropy is negative, again in order to minimize the free energy, the liquid crystal molecules will align perpendicular to the direction of the electric field.

$$f = -\int \vec{D} \cdot d\vec{E} = -\frac{1}{2} \left[ \varepsilon_\perp E^2 + (\varepsilon_\parallel - \varepsilon_\perp)(\hat{n}.\vec{E})^2 \right]$$  \hspace{1cm} (2.1.3.1.3)
$$\vec{D} = \varepsilon_\bot \vec{E} + (\varepsilon_\parallel - \varepsilon_\bot)(\hat{n} \cdot \vec{E})\hat{n}$$  

(2.1.3.1.4)

Figure 2.3. Response to the applied electric field for liquid crystal molecules having positive and negative dielectric anisotropy. Reproduced from [1], p.4.

**2.1.3.2 Liquid Crystal/Solid Surface Interaction**

Liquid crystal cells have two glass plates with the liquid crystal material held in between. The thickness of these cells is in the order of micrometers and is negligible compared to the area of the glass plates. Thus the interaction of the liquid crystal material with the glass surface, often coated with a transparent electrode is of significance [4]. The liquid crystal molecules directly in contact with the solid surface is affected most which changes their physical properties. However the liquid crystal molecules flow like a liquid. Thus the effect of the surface liquid crystal interaction on the bulk molecules will depend on the distance from the surface as well as the direction [5].
2.1.3.3 Liquid Crystals Alignment

The alignment of liquid crystals is of fundamental importance. In this section I discuss the alignment of liquid crystals due to the solid surface properties. Two important parameters which determine the liquid crystal alignment are the liquid crystal /air surface tension $\gamma_{L/A}$ and the solid/air surface tension $\gamma_{S/A}$. When a liquid crystal molecule is placed on the solid surface, the balance between these two surface tensions determines the alignment of the liquid crystal molecules such that the surface energy is minimized. As a good rule of thumb, in a situation where $\gamma_{L/A} > \gamma_{S/A}$, the liquid crystal alignment is homeotropic which means that the rod-like molecules align perpendicular to the surface. When $\gamma_{L/A} < \gamma_{S/A}$ the liquid crystal alignment is planar which means that the liquid crystal molecules align along the surface. In the scenario that $\gamma_{L/A} \approx \gamma_{SA}$ the liquid crystal molecules tend to take up an alignment which is tilted [6]. The three different alignments have been shown in fig. 2.4.

The tilted alignment is often produced by light rubbing of the PI coating. At thermal equilibrium, the liquid crystal molecules align based on the rubbing direction so as to minimize the surface energy [7-9]. This is called the easy axis as shown in fig. 2.5. The easy axis has polar and the azimuthal angles represented by $\theta_0$ and $\phi_0$ respectively in fig. 2.5. In case of homeotropic alignment, $\theta_0 = 0^0$ and $\phi_0 = 0^0$. In case of planar alignment $\theta_0 = 90^0$ and $\phi_0$ is fixed or arbitrary. The director is denoted by the polar angle $\theta$ and the azimuthal angle $\phi$.

The director and the easy axis are correlated through the extrapolation length. The extrapolation length is the distance from the surface where the director meets the easy axis. The extrapolation
length given by $K/W$, where $K$ is an elastic constant and $W$ the surface anchoring coefficient. When the anchoring is strong, the extrapolation length is zero. However when the anchoring is very weak, the extrapolation length is large.

Figure 2.4. Liquid crystal between two substrates having different alignment (a) Planar alignment (b) Homeotropic alignment (c) Tilted alignment. Reproduced from [1], p.7.

Figure 2.5. Schematic of the easy axis and the director at the liquid crystal solid surface interface. Reproduced from [1], p.6.
2.1.4 Optics of Liquid Crystals

The anisotropic shape of nematic liquid crystal molecules provides several physical properties which are dependent on direction. One such property is the refractive index. When the direction of polarization of light is parallel to the optic axis of the molecule, the measured refractive index is called the extraordinary refractive index $n_e$. On the other hand when the polarization of light is perpendicular to the optic axis of the molecules, the measured refractive index is the ordinary refractive index $n_o$. This is shown in fig. 2.6. As shown in the figure, $n_o < n_e$. For any other direction in between, the refractive index is given by the effective refractive index $n_{eff}$. The effective refractive index is given by eq. 2.1.4.1. The angle $\theta$ is the angle between the wave vector $\hat{k}$ and the optic axis of the molecules. The wavevector $\hat{k}$ which provides the direction of propagation of light is perpendicular to the direction of the polarization of light.

The liquid crystal molecules can be controlled by an electric field and made to turn in order to control the refractive index. This has many electro-optical applications [10-19]. When the electric field is applied, the molecules turn when the voltage is above a threshold voltage. This is shown in fig. 2.6.
Apart from homogenous electric field, inhomogeneous electric field can also be generated using different techniques. This results in non-uniform distributions of the refractive index profile. The various methods popularly used are electrical addressing of multiple electrodes [20], fresnel-type lenses filled with LCs [21-23], patterning of a solid electrode either slit type [24-26], or hole type [27] and other approaches [28].

2.2 Cylindrically symmetric inhomogeneous electric field

Cell configurations used to create symmetric inhomogeneous electric field, reported so far, utilize different cell configurations [29,30]. This is shown in fig. 2.7. A general review of important literature papers each representing one of these configurations has been presented below.
2.2.1 Zones

Inhomogeneity in the electric field across the cell region can be created by electrodes having concentric ring structures called zones [31,32]. In the simplest case, a set of alternate electrode rings may be grouped together and provided with the same voltage, resulting in reorientation of the LC molecules stacked above it. As an example, a TN cell, having alternate electrode ring structures, is shown in fig. 2.8.
As seen in fig. 2.8, the refractive index of the LC material stacked above the zones alternate and therefore the optical phenomenon that controls the transmission of light through the zones, is diffraction. The width of the zones can be uniform or non-uniform. In the first case, when the zone width is uniform, i.e the periodicity is fixed, the cell works as a regular diffraction grating. This is shown in fig. 2.9.

![Diffraction Grating Schematic](image)

Figure. 2.9. Schematics of a diffraction grating with fixed periodicity [31].

In case of a diffraction grating,

\[ d \sin \theta = n \lambda \]  

(2.2.1.1)

where \( d \) is the period of the grating. For first order, \( n = 1 \), and hence

\[ \sin \theta = \frac{\lambda}{d} \]  

(2.2.1.2)
Let us now suppose that a zone plate is formed of diffraction grating of the same periodicity. Then, as shown in fig. 2.10, the first order diffraction for a pair of slits at a distance \( r \) from the optical axis of the zone will be at a distance \( z \) from the plate. Hence,

\[
\tan \theta = \frac{r}{z} \tag{2.2.1.3}
\]

Comparing, eqns. 1.2.1.2 and 1.2.1.3, and taking the paraxial approximation into account, such that \( \sin \theta \approx \tan \theta \), I get

\[
\tan \theta = \frac{\lambda}{d} = \frac{r}{z} \tag{2.2.1.4}
\]

So,

\[
z = \frac{(r \times d)}{\lambda} \tag{1.2.1.5}
\]

Figure. 2.10. Zone plate as a diffraction grating of uniform periodicity [31].

Now, for a zone plate, as shown in fig. 2.10, with each successive zone from the center, \( r \) increases. If \( d \) is a constant, \( z \) will have different values for each order of the diffraction grating. Based on the grating period \( d \), different values of \( z \) can be obtained. As a special case when
there is a decreasing periodicity, and the radius of any zone is related to the first zone in given by eq. 2.2.1.6 [32].

\[ r_n^2 = n \cdot r_1^2 \]  

(2.2.1.6)

Where, \( r_n \) is the radius of the \( n^{th} \) zone, and \( r_1 \) is the radius of the first zone, as shown in fig. 2.11, the entire first order light from each of the zones, collect at the same point. This is called a zone plate.

![Diagram of a Fresnel zone plate](image)

Figure. 2.11. Two dimensional view of a Fresnel zone plate [32].

The focal length of the zone plate is given by eq. 2.2.1.7 [32].

\[ f = \frac{r_1^2}{\lambda} \]  

(2.2.1.7)

where, \( f \) is the focal length, \( r_1 \) is the radius of the \( 1^{st} \) zone and \( \lambda \) is the wavelength.

Usually the fabrication of a zone plate is complicated, with feature size of even 1\( \mu \)m reported and the electrical connections involve several fabrication steps. Also as reported in the literature, the zones are further divided into sub-zones in order to digitize the phase profile. The sub-zones of different zones are shunted together by electrodes. Each of these shunted sub-zones are provided the same voltage. Hence, each of the shunted subzones have the same phase profile in different zones.
This results in the discretization of the phase and is the beginning of the digital optics. The focal length obtained as a consequence of the phase profile can be varied by electrically changing the number of subzones. As can be seen from eq. 2.2.1.7, the focal length can be reduced by dividing any zone into many sub-zones. Zone plates are optimized to achieve high diffraction efficiency, fast switching and low driving voltages. The diffraction efficiency is the fraction of output intensity to total intensity for a given order. The diffraction efficiency is obtained as function of the phase level. This is given by eq. 2.2.1.8 [32].

\[
\eta = \text{sinc}^2 \left( \frac{1}{P} \right) = \left[ \sin \left( \frac{\pi}{P} \right) / \left( \frac{\pi}{P} \right) \right]^2
\]

(2.2.1.8)

where, \( \eta \) is the the diffraction efficiency and \( P \) is the phase level. It is derived using the transfer function of a Fresnel zone plate. The transfer function is obtained using fourier optics.

In Fresnel zone plate configurations with stepped phase profiles, a very high diffraction efficiency of 97.7% can be achieved. A low driving voltage of 2.5V, a very large aperture size of 10mm and a small cell gap of 3\( \mu \)m has been reported. Large focal length tunability from 6.6cm to 40cm has been reported corresponding to the same dimensions. In spite of the many fabrication steps involved, these optical elements used as lenses have found ophthalmological applications. Although these have many advantages, a major problem with these type of lens is that they are polarization dependent and may require orthogonal stacking [32].

A polarization independent Fresnel zone plate has also been reported [33]. The construction involves alternating orthogonal photoalignment using linear polarized UV(LPUV). Initially the
entire PI layer is photoaligned using LPUV. Then by using a mask the exposed areas are realigned in the orthogonal direction with respect to the first alignment. The diffraction efficiency of the device is electrically controllable and is maximum when the phase shift of the two corresponding zones is \( \pi \). The diffraction efficiency for first order diffraction is given by eq. 2.2.1.9 [33].

\[
\eta = \text{sinc}^2 \left( -\frac{1}{2} \right) = \left[ \frac{\sin \left( -\frac{\pi}{2} \right) / \left\{ -\pi \right\}}{\left\{ -\pi \right\}} \right]^2
\]  

(2.2.1.9)

As reported in the literature, a phase shift of \( \pi \) between odd and even zones occurs at 0V for a 4.1\( \mu \)m cell and at 1V for 7.5\( \mu \)m cell. An aperture size of 7.2mm has been reported with 80 zones within the aperture region. The radius of the innermost zone is 403.8\( \mu \)m. One major drawback of this lens is that the diffraction efficiency is quite low and is \( \sim 41\% \). The focal length tunability is between 30 cm and \( \infty \).Improvisation of the above mentioned zone plate types have been reported [34-37]. They include lenses which are both polarization dependent as well as polarization independent and have varying range of tunability in focal length.

2.2.2 Hybrid type

Another way to create an inhomogeneous electric field is to use a curved electrode. A form of curved electrode is an ITO coated shell. This is shown in fig. 2.12. Such a configuration is called a hybrid type configuration [38-47]. The setup consists of a glass shell, coated from inside with ITO and laminated over a LC cell, in which the bottom substrate is uniformly coated with the electrode from inside. The top substrate is partially coated so as to ensure continuity in the electrode connections between the glass shell and the top substrate. The shell can be filled with air or polymer each having their own applications. The inhomogeneous electric field in
conjunction with the LC material creates a refractive index profile which creates a lensing effect. A lens aperture of 6mm and a shell thickness of 200μm has been reported [47]. The thickness of the glass slab \(d_g\), forming the top and bottom substrates, is 550μm. The distance \(d_s\) is equal to 720μm. The thickness of the cell is 25μm. The LC alignment is planar, with anti-parallel rubbing and with a pretilt angle of 6°. The curvature of the glass shell produces an inhomogeneous electric field, which causes a refractive index profile producing a lensing effect.

![Figure 2.12. Schematic of a hybrid LC lens setup as reported. Reproduced from [47], p.11293.](image)

The focal length \(f\) of the lens will then be given by eq. 1.2.2.1 [47].

\[
f = \frac{r_0^2}{2d(n_e - n_{effective})}
\]  

(2.2.2.1)

Where, \(r_0\) is the radius of the aperture, \(d\) is the cell thickness and \((n_e - n_{effective})\) is the difference in refractive index between the centre and the edge. When the voltage is above the saturation value, the LC molecules at both the centre and the edge get aligned with the electric field. The refractive index at the center and the edge is both \(n_0\) and the focal length is \(\infty\).
The focal length can be maximized by maximizing the value \( n_e - n_{\text{effective}} \). For that to happen, the voltage at the centre should be lower than the threshold value such that the LC molecules are planar. The refractive index at the center will then be \( n_e \). The refractive index at the edge of the hole region should get completely aligned with the direction of the electric field, such that, \( n_{\text{effective}} \) becomes equal to \( n_0 \). In that situation, the value \( (n_e - n_{\text{effective}}) \) will simply be equal to the birefringence \( \Delta n \).

\[
\Delta n = (n_e - n_0)
\]

(2.2.2.2)

Since the orientation of the LC molecules is affected by the electric field, the parameters that determine the electric field, has been investigated. A model analogous to the capacitance model has been suggested. Accordingly,

\[
E_{\text{Center}} = \frac{V/\varepsilon_{LC}}{(d_{LC}/\varepsilon_{LC})+(d_g/\varepsilon_g)+(d_s/\varepsilon_m)}
\]

(2.2.2.3)

\[
E_{\text{Edge}} = \frac{V/\varepsilon_{LC}}{(d_{LC}/\varepsilon_{LC})+(d_g/\varepsilon_g)}
\]

(2.2.2.4)

where as shown in fig. 2.12, \( V \) is the applied voltage, \( d_s \) is the distance between the upper plate and the apex of the shell along the optic axis, \( d_g \) is the thickness of the glass plate and \( d_{LC} \) is the thickness of the LC cell. \( \varepsilon_{LC}, \varepsilon_g \) and \( \varepsilon_m \) represent the dielectric constant of the liquid crystal, glass and the material used to fill the shell. A couple of interesting observations can be made from eq. 2.2.2.4. As the dielectric constant of the material filling the shell is decreased, the electric field at the center of the shell decreases as well. Since the dielectric constant of air is less than the dielectric constant of polymer,
it is preferable to have an air filled glass shell as compared to a polymer filled glass shell while
trying to reduce the electric field at the center.

Also as seen from 2.2.2.3 and 2.2.2.4, the electric field is affected by the ratio, \( (d_{\text{LC}}/\varepsilon_{\text{LC}}) : (d_g/\varepsilon_g) \).

It has been reported that lowering the ratio from 27 to 3, the applied voltage reduces from 140 \( V_{\text{rms}} \) to 20 \( V_{\text{rms}} \). This is possible by using a thinner glass substrate and a thicker cell.

There are certain consequences of a thicker cell. One of the consequences is a shorter focal length as seen from eq. 2.2.2.5. Also a thicker cell leads to a higher relaxation time

\[
\tau_{\text{decay}} = \frac{\gamma_1 d^2}{K_{33} \pi^2}
\]  

(2.2.2.5)

where \( \tau_{\text{decay}} \) is the relaxation time, \( \gamma_1 \) is the rotational viscosity and \( K_{33} \) is the bend elastic constant. As reported, for the setup shown in fig. 2.12, when the thickness of the LC cell is doubled, from 25\( \mu \)m to 50\( \mu \)m, the focal length reduces to half, from 96cm to 48cm. The relaxation time as expected from eq. 2.2.2.5, increases from 1sec to 4sec. Increasing the thickness, in order to reduce the focal length leads to higher relaxation time.

As seen from eq. 2.2.2.1 and also mentioned earlier, the focal length is also dependent on the birefringence of the LC material. Hence using a LC material of higher birefringence may help to reduce the focal length. Optimizing all these parameters, like higher cell thickness, a 1:3 ratio between the glass and the cell thickness and by using a conductive polymer having a refractive index similar to glass leads to a larger focal length tunability varying between 15.4cm and \( \infty \) [48]. The aperture in this setup is around 4.2mm. The results reported therefore validate the models discussed above.
Improvisation of similar setup and by using a plastic Fresnel lens along with a dual frequency LC, helps to create a lens with a larger lens diameter. As reported, the focal length of such a lens can vary from -1.5 D to 1.2 D [49].

2.2.3 Hole patterned electrode

Instead of a curved electrode, a simple way to create an inhomogeneous electric field is to use the flat patterned electrode [50-61]. One of the patterns can be that of a hole in the electrode coated substrate. The hole creates an inhomogeneous electric field and the LC directors get realigned with the electric field. This helps to create a refractive index profile creating a lensing effect. The hole patterned lens forms a very attractive option from an application point of view because it involves some simple processes and can qualify as a flat adaptive lens. The processes involve etching of the ITO and can be carried out in the minimum number of steps in any clean room environment. A typical schematics showing the arrangement of the LC molecules inside the cell with and without the application of voltage is seen in fig. 2.13
Figure 2.13. Hole patterned LC lens with homeotropic alignment (a) without application of voltage (b) with application of voltage.

In fig. 2.15(b), at the center of the cell, the LC molecules are perpendicular to the polarization of the incoming light and hence the refractive index is $n_o$. At the edge of the hole, the LC molecules are at an angle to the polarization of incoming light and hence the refractive index is $n_{\text{effective}}$. The value of $n_{\text{effective}}$ here is less than $n_e$. 
The important parameters for any hole patterned lens are the, focal length, the hole aperture and the driving voltage. Microlens with focal length of 6mm, aperture size of 750μm have reported a low driving voltage of 4V [62]. For a hole patterned lens, with a hole etched out in one of the surface electrodes, the ratio of the hole diameter: the cell thickness should have been experimentally found to be around 2-3, in order to obtain ideal optical properties [Ch1 13]. The cell thicknesses are usually in the micrometer range. That would allow a hole diameter also in the micrometer range. Larger lens aperture, based on a modal control approach has been reported. The lens has a hole diameter of 7mm, a minimum focal length of 76 cm and a driving voltage of 52V_{rms}. All hole patterned lenses reported so far have been found to be using AC voltages [63-65].

2.3 Motivation and goals

Various LC cells with electrode patterns to create inhomogeneous electric field, reported so far, use AC as the driving voltage. The electro-optic response of these cells have been measured quantified and reported by various authors. The electro-optic response of these cells using DC voltage has not been reported yet. Differences in electro-optic response between AC and DC voltages, for LC cells without patterning and having continuous electrodes have been reported in the literature [66,67]. However no such study exits for patterned electrodes which create an inhomogeneous electric field. This makes comparison of the electro-optic response on the application of AC and DC voltages for patterned LC cells an interesting study. Amongst the
patterned electrodes, the simplest way to create a pattern is a hole patterned electrode. This requires the least number of fabrication steps to create the cell. The thesis therefore uses a hole patterned LC cell, to compare the electro-optic response for samples, using AC and DC voltages. Also the starting hole diameter is 1mm and studies on 2mm, 3mm and 4mm hole diameters have also been reported. The 2mm hole diameter is the size used in many cellphone camera applications [68].

2.4 Summary

Optical elements using patterned cells and using LC which helps to create inhomogeneous refractive index profile have been studied extensively and are focused in this thesis. The electro-optical responses of cells with various kinds of patterned electrodes, producing inhomogeneous electric field have been reported in literature. The various patterns have been further classified in terms of zone, hybrid and hole pattern. In terms of fabrication, hole patterned cells have been found to be relatively simple. All patterned cells reported so far use AC voltage. This is because it is well understood that, in the absence of electrical contaminants, the response of the liquid crystal to AC and DC voltages will be the same. This would indicate that the response of patterned cells should be the same regardless of the frequency of the applied voltage. However, as reported below, I observed a significant difference between the observed responses using the two systems. This, to our knowledge, is the first observation of such a phenomena in this area of research. As such, it is not understood. The observation of this effect and its origin are the main topic of this thesis.

In order to carry out a more in-depth analysis, a flow chart, as shown in fig. 2.14, helps to explain the experimental setup and detection used in the subsequent chapters. A detail of the cell
preparation has been discussed in Chapter 3 and the characterization results and analysis has been discussed in Chapter 4 respectively.

Figure. 2.14. Processes involved in the fabrication and characterization of hole patterned LC cells.
CHAPTER 3

Fabrication of hole patterned liquid crystal cells

3.1 Introduction

The fabrication of hole patterned LC cells has been discussed in this chapter. The fabrication involves several clean room processes. These include glass cutting, acid etching of the substrates, spin coating of the alignment material, rubbing and gluing of the substrates. Finally the substrates have to be assembled and individual cells have to be extracted. All the necessary steps required to create a LC cell has been discussed in this chapter. Also, any electro-optic use of these cells requires filling the cells with LC material and the option to access the electrodes on both the substrates. Details regarding them have also been provided.

3.2 PI alignment layer

Most hole patterned liquid crystal cell configuration, uses a PI which provides a planar alignment [Ch1 84-86]. However, because of the alignment, the optics of the cell is still dependent on the polarization of light [1,2]. The polarization dependence in the hole region can be eliminated by using homeotropic liquid crystal configurations with a material of negative or positive dielectric anisotropy [3]. Studies of the electro-optic properties with hole patterned LC cells, having homeotropic alignment, have been reported and provides the possibility to use PI with homeotropic alignment for the fabrication of our cells [4].
Based on the availability of PI material which provides a homeotropic alignment, we have chosen SE-1211, 6% by weight dissolved in a solvent. SE-1211 has been reported as an excellent homeotropic alignment layer and is well characterized [5,6]. The solvent in which it is dissolved has a composition of 54% N-Methyl-2-Pyrrolidone and 40% Butylcellosolve. The PI can be diluted further using Solvent 26 which consists of 60% N-Methyl-2-Pyrrolidone and 40% Butylcellosolve. The PI is already polymerized and does not require any further polymerization. It is characterized by long alkyl side chains and an aromatic backbone. The side chains promote homeotropic alignment, while the backbone provides planar alignment. In order to use the PI as an alignment layer, the solvent is dried off. In our experiments, the drying of the solvent is done at 180°C overnight. Drying above 180°C imidizes the backbone, cleaves away a fraction of the side chains and facilitates a less homeotropic alignment [7].

3.3 Cell preparation

3.3.1 Glass cutting

An ITO coated glass plate, 7” x 7”, having an ITO resistance 330 Ω/cm² is cut out into smaller pieces of 3.5”x 3.5”. The cutting is done on the table using the glass cutter manually. The glass is first cleaned with ethanol and acetone. The side without the ITO is marked out symbolically with a letter ‘R’, using the sharpie, at the four corners. This can be seen in fig. 3.1. The ITO coated surface is placed on the top and the cutting is done over the ITO coated surface. Air blower is used after cutting to clean up the glass shreds. The final cleaning is done using
acetone and ethanol again. Gloves can be used at the time of cleaning to prevent any finger prints on glass. The glass plates are then stored using a boat for the next stage of the process.

![Image showing initial cutting of glass](image)

**Figure. 3.1.** Initial cutting of the glass into four equal slabs of 7”x7”. The marking ‘R’ designates the non-ITO coated surface.

### 3.3.2 Mask pattern using sticker cutter

In order to acid etch the ITO, the region in the glass plate that should not be exposed to the acid is protected using a mask. The mask consists of a black vinyl sticker made out of plastic. This sticker mask is selected based on the adhesion strength. Using the Corel Draw 12, the pattern is first drawn out. In our case this pattern is a hole. The size of the hole can be varied using the software. The pattern is then printed out on a plastic sheet using the laser cutter. In order to make a comparative analysis different hole sizes have been selected. The different sizes of the hole diameters selected are equal to 1mm, 2mm, 3mm and 4 mm.
3.3.3 Lamination of the mask on the substrate

The mask of suitable size, little less than the glass plate is laminated on to the ITO coated side of the substrate. The lamination is carried out inside a clean room environment. A transfer sticker is used in the lamination process. The transfer sticker is first laminated on to the mask using a roller. Using a sharp blade the mask along with the transfer sticker is cut out along the edges. The paper attached to the mask is then removed while the mask along with the transfer sticker is laminated to the glass substrate. Care needs to be taken during the lamination, such that the entire mask lies within the glass substrate. The transfer sticker is then removed. In order to ensure that no over etching occurs during the acid etching process, it is necessary that the glue at the edges of the hole in the mask, sticks firmly to the glass plate. Thus, to achieve excellent etching and high yield, the edges of hole in the mask are manually pressed against the glass substrate. The glass substrate is now ready for etching. This is shown in fig. 3.2 which shows one of the glass substrates which have been laminated with an acid-resistant sticker and is ready for acid-etching. The other substrate is without any lamination.
Figure. 3.2. Schematics of two glass substrates to prepare cells, one laminated with the acid-resistant sticker and the other without any lamination.

3.3.4 Etching of the ITO in acid solution

The acid solution used for the etching of the ITO comprises of 38.81 % HCl, 3.88% HNO₃ and 57.3% H₂O. The acid is kept inside an acid bath. The substrate is first mounted on an acid resistant boat and is dipped slowly into the acid bath. It is important to open the lid of the acid bath using acid resistant gloves. The handle of the boat should be out of the acid. The etching time is optimized and 50 minutes is found to be the optimum time required for etching. As shown in fig. 3.3, very low etching time leads to underetching at the edges of the hole and the resistance measured inside the hole region is low. Similarly very high etching time leads to overetching. An etching time of 50 min provides a very well defined hole edge and is used for fabrication of the samples. Once the etching is done, the boat is taken out slowly and is lowered.
into a water bath for a minute. The boat is next cleaned with tap water to remove any residual acid solution. Further cleaning is carried out using deionized water to remove any residual salt deposited by tap water. The mask is removed and the substrate is ready for the next step.

![Graph showing resistance vs. etching time](image)

Figure 3.3. Resistance close to the edge of the hole region and inside the hole as a function of etching time.

### 3.3.5 Spin coating the PI layer on the substrate

Spin coating the PI solution on to the substrate, first requires adequate cleaning of the substrate. A chuck of proper size is mounted in the spin coater. The vacuum is turned on and the substrate is placed on to the spin coater. From the software, the spin speed and the spin time is controlled. The spin speed used in our experiments is 1500 rpm for 30 sec.

The PI solution is dispensed in a way such that it just covers little less than the entire substrate. In situations when the solution is right up to the edge, during the spinning process, the excess solution tends to get in between the chuck and the substrate. It is also advisable to clean out the excess PI
before evaporating out the solvent using an oven. A coating thickness of 1 μm using PI: Solvent 26 ratio of 1:0 could be achieved in our experiments. The final configuration can be seen in fig. 3.4.

![Diagram of two substrates coated with PI. The upper substrate has ITO etched regions while the lower substrate has uniform ITO coating. Many different cells are made out from each substrate.](image)

3.3.6 Evaporating the excess solvent from the PI

To remove the excess solvent from the PI, the PI coated substrate is first dried in a hot plate for 1 min at 90°C. It is further evaporated using a preheated oven. Before inserting the PI coated substrate in the wet condition, the oven is cleaned to ensure that particles from the roof and walls of the oven do not settle on to the substrate. Once the oven is cleaned, the temperature of the oven is maintained for 2 hrs at 180°C before the substrate is thrown into the oven for drying. The recommended time by the manufacturers [8] is 30 min. The oven is kept switched on for 30 min and the sample is kept in the oven overnight. For SE-1211, the PI used in our experiments, a temperature above 180°C leads to cleaving of the alkyl chains and hence planarization [8].
3.3.7 Printing the glue pattern on the substrate mixed with spacers

To assemble two substrates and create a cell, dispensing of the glue is required. The glue is dispensed over one of the substrates. In our case the substrates over which the glue is dispensed is the patterned substrate. The spacers which determine the thickness of the cell is mixed with the glue. The spacers used in our experiments are glass spheres of 50μm diameter. The glue pattern is designed using Corel Draw 12 and is processed to make it compatible with Asymtek Glue dispenser. A trial run using a thick sharpie as the needle is carried out on the glue dispensing table. This helps in the alignment and also helps to determine the uniformity of the coating. In case of lack of uniformity, the coating table has to be adjusted, even though such adjustments are rarely required. The glue syringe is now loaded and is connected to the vacuum. Using the software, the drag time is adjusted. Next, the height of the needle is adjusted at the center of the substrate. Dispensing glues is analogous to laying a rope and the height of the needle needs to be optimized in order to have a proper dispensing.
3.3.8 **Baking the glue**

The dispensed glue is baked using an oven. Depending on the freshness of the glue the baking time may vary between 25 minutes to 45 minutes. Usually fresh glue requires more time to bake. The temperature of the oven is kept at 70°C.

3.3.9 **Assembling the cell using the T-shirt press**

Once the glue is baked, the substrate with the glue coated side is kept on top of the T-shirt press. The other substrate, with the coated sides of the two substrates facing each other, is gently placed on top of it. A silk cloth is spread on top of the assembled substrates and the pressure in the T-shirt press is maintained at 60 psi. High temperature is attained in the T-shirt press and usually takes 10-12 hrs for the assembled substrates to attain normal room temperature.

3.3.10 **Cutting the plate using glass cutter to prepare individual cells**

The individual cells to be used for experiments, is extracted out of the assembled substrates. The cells are cut manually using the glass cutter. The cutting is done by scribing on the cells orthogonally. The extracted cell has access to both the ITO coated substrates and hence the electrodes.
3.4 LC material

The choice of the LC material is done so as to achieve maximum reorientation of the molecules, under the influence of the electric field. The initial alignment of the LC molecules as provided by the alignment layer is homeotropic. Homeotropic alignment has been chosen for reasons mentioned before. In order to get maximum refractive index distribution between the center of the hole region and the edge of the hole on the application of voltage, a LC material of negative dielectric anisotropy is selected. The dielectric anisotropy of the LC material is -5. This ensures that on application of high enough electric field, the molecules outside the hole region is planar, while those at the centre of the hole region remains homoetropic. The birefringence of the LC material is 0.15.

3.5 Filling of the liquid crystal cell and applying the electrodes

Once individual cells have been cut out, the polyimide in the exposed ITO regions is removed. This can be done using NMP. The electrodes are then laminated on to the exposed ITO regions. The electrode used in our experiments is a threaded copper electrode. The filling of the cell with the LC material is then done while keeping the cell on a hot plate. The hot plate reduces the viscosity of the LC material, allowing faster filling. Once filled, the cell is sealed and is allowed to relax for at least 24hrs. This is to ensure that the LC material inside the cell attains the homoetropic alignment imposed by the PI alignment layer. The final cell after filling and without any application of voltage is as shown in fig. 3.13 (a), shown earlier.
3.6 PI thicknesses

The PI:Solvent used in our cell preparation are 1:1, 1:2, 1:4, 1:8 and 1:16. Thickness for samples with various PI:Solvent ratio have been provided in the literature [9]. This is shown in fig. 3.5

![Diagram showing PI thicknesses for different ratios](image)

Figure. 3.5. Thicknesses of different PI used in the experiments [9].

3.7 Summary

A LC cell has been fabricated and filled with the LC material. The steps involved in the fabrication have been mentioned in details. The choices of the PI and the LC material have also been discussed. Electro-optic experiments have been performed using these cells and have been discussed in subsequent chapters.
CHAPTER 4

Experimental setup, results and discussion

4.1 Introduction

Hole patterned LC cells having different hole diameters of the electrode etched region have been fabricated. As mentioned in the previous chapter, the hole patterning is done by etching one of the two ITO coated substrates. The hole apertures reported in the literature vary between 300 μm to 7 mm [1, Ch1 65]. The hole patterned LC cells reported so far utilize AC voltage. It has been reported that the use of DC voltage in LC electro-optical devices has the possibility to alter the electro-optical behavior of the devices [2,3]. However, the electro-optical properties of the hole patterned LC cell on the application of the DC voltage has not been reported yet. In this research, various electro-optic properties of hole patterned LC cells, on the application of AC and DC voltage for the same cell has been compared and investigated. Cells with different hole sizes 1mm, 2mm, 3mm and 4mm in diameter, have been fabricated and characterized for their optical properties, with a cell thickness of ~50 μm being kept constant. Hole size of ~2mm has particular importance in the applications related to cell phone cameras lenses [4, Ch1 68]. The etching is carried out using an acid-resistant sticker as mask. The experimental setup for characterization of the cell has been described below.
4.1.1 Experimental setup for characterization

The hole patterned cell creates an inhomogeneous electric field on the application of voltage. This inhomogeneous electric field creates a refractive index profile. This refractive index profile can lead to phase modulation of light [5]. Interferometric technique is often used to measure phase modulation. A interferometric setup to characterize hole patterned cells using polarized light has been provided in ref [6]. The cell is characterized by the interference pattern produced as a result of the inhomogeneous electric field created inside the hole by the application of voltage. Similar interferometric setup using a polarized optical microscope with the cell placed between crossed polarizers have also been reported [Ch3 4]. An optical microscope setup, where the cell can be placed between crossed polarizers, with the optics as shown in fig. 4.1, is hence used for characterization of our hole patterned LC cells. The cell is kept on the stage of a transmission microscope and between two crossed polarizers. A white light source is used in the experimental setup. The eyepiece is replaced with an Olympus camera to enable collection of images.
Figure 4.1. Schematics of an optical transmission polarizing microscope with the sample kept in between crossed polarizers and on the specimen plate. Reproduced from [7], fig. 9.

The initial alignment of the LC cell produced by the polyimide is homeotropic and when seen under crossed polarizers, without application of any voltage, the image appears dark. As voltage is applied, an electric field is created, which helps to reorient the LC molecules resulting in enhanced transmission. The re-orientation happens only above a critical electric field. Images of both inside and outside the hole etched region is taken and analyzed further.

Both AC and DC voltages are applied to the cell in the course of the experiment. For AC voltages, frequency of 60Hz have been used. The texture of the hole region have been found to
be the same for higher frequencies. Hence only the results for a frequency of 60Hz have been presented.

4.2 Results and discussion

The results obtained through the characterization of hole patterned LC cells have been analyzed. Electro-optic response of hole patterned LC cells, starting from cells with initial homeotropic alignment have been reported using a LC material of both positive as well as negative dielectric anisotropy and using AC voltage [Ch3 1,Ch3 4]. In our case the electro-optic response using both AC as well as DC voltages have been compared for a LC cell with an hole etched electrode region and with a particular hole diameter. The comparison is done both inside and outside the hole etched electrode region. Electro-optic response has been reported in the literature for cells with unpatterned electrodes on both the substrates. This is the same as the region in our cells outside the hole etched electrode region. While considering the electro-optic response of our cells outside the hole etched region, for both AC as well as DC voltages, our aim is to compare it with those reported in the literature. The electro-optic response inside the hole etched region, using the AC voltage, has been reported before. However, using the DC voltage, the electro-optic response is being reported, for the first time, in this thesis. This is then repeated for cells with different diameters for the hole etched region. Four different hole sizes for the hole etched electrode region have been considered in our study. The diameter ranges from 1mm to 4mm.
4.3 Re-orientation of the LC molecules using the electric field

The electric field acts on the dipole moment of a single LC molecule producing a torque. This is given by eq. 4.3.1.

\[ \vec{\tau} = \vec{\mu}_{molec} \times \vec{E} \]  

(4.3.1)

where, \( \vec{\tau} \) is the torque, \( \vec{\mu}_{molec} \) is the dipole moment and \( \vec{E} \) is the electric field.

For the bulk material, the average of all the individual dipole moments of the LC molecules is given by the electric polarization \( \vec{P} \)

\[ \vec{P} = \rho < \vec{\mu}_{molec} > \]  

(4.3.2)

where, \( \vec{P} \) is the electric polarization which is the dipole moment per unit volume, \( \rho \) is the density and \( < \vec{\mu}_{molec} > \) is the average dipole moment.

The electric displacement is then given by

\[ \vec{D} = \varepsilon_0 \vec{E} + \vec{P} \]  

(4.3.3)

where, \( \vec{D} \) is the electric displacement, \( \varepsilon_0 \) is the vacuum permittivity and \( \vec{P} \) is the electric polarization.

The torque experienced by the bulk LC molecules due to the electric field is then given by \( \vec{\tau}_{total} \) and is given by eq. 4.3.4

\[ \vec{\tau}_{total} = -\vec{D} \times \vec{E} \]  

(4.3.4)

This torque is balanced by other competing torques like the elastic torque. The balance of all these torques, determines the final orientation of the LC molecules.
The critical electric field and voltage required to re-orient the LC molecules, is obtained by the minimization of the free energy density. [8]. The critical value of the electric field is

\[ E = \frac{\pi \sqrt{\frac{K}{\varepsilon_0 |\Delta \varepsilon|}}}{d} \]

(4.3.5)

\[ V = \frac{\pi \sqrt{\frac{K}{\varepsilon_0 |\Delta \varepsilon|}}}{d} \]

(4.3.6)

The voltage called the Fréedericksz distortion voltage is given by eq. 4.3.6. This voltage is valid for both the AC as well as the DC voltages. However, in-order to match with experimental values, additional factors like the presence of impurities have to be considered.

The LC molecules outside the hole patterned region is subjected to uniform electric field. This changes inside the hole patterned region where inhomogeneous electric field is produced. In the next section, the inhomogeneous electric field due to hole patterned electrodes and the corresponding electro-optic phenomenon associated with have been discussed.

### 4.3.1 Inhomogeneous electric field due to hole patterned electrode

Hole patterned electrodes have been reported to create cylindrically symmetric inhomogeneous electric field in LC cells with the application of voltage [9]. The inhomogeneous electric field creates a refractive index profile. When a polarized light is transmitted through the hole region of such a LC cell, due to the presence of the refractive index profile, an interference pattern is observed. The interference pattern can be confirmed through the presence of alternate bright and dark rings.
A design which creates an inhomogeneous electric field for cells with large hole etched region has been reported by a Japanese group [Ch1 86]. It uses a hole patterned electrode in one of the substrates and an unpatterned electrode on the other substrate. In this design, there is a dielectric layer between the patterned electrode and the LC layer. Inhomogeneous electric field, goes all the way from the edge to the center of the hole patterned region for appropriate voltages. In this way it is possible to create a hole pattern of large sizes, depending upon the applied voltage. The design parameters, the experimental setup and the corresponding image of the hole region using polarized light have been provided below.

**Hole diameter:** 7 mm

**Design:** Fig. 3.2 shows the schematic of the cell design. There is only one Aluminium electrode. The glass slab between the Aluminium electrode and the LC has a thickness of 1.3 mm. The thickness of the LC layer is 130 μm. The LC used has \( \Delta \varepsilon = 12.3 \).

**Experimental setup:** The experimental setup is as shown in ref [6]. The LC cell is kept between a polarizer and an analyzer which are at right angles with each other. The rubbing direction of the cell is at 45\(^\circ\) to each of the polarizer. A function generator provides the voltage to the cell.

**Image:** The image of the hole etched region is obtained with the cell placed between crossed polarizers. In this design, a suitable voltage is first applied outside the hole region which creates the inhomogeneous electric field. This we define as the “applied voltage”. The “applied voltage” is 52V AC. A stopper of diameter 5 mm is used to obtain the image. This is shown in fig. 4.3.
Figure. 4.2. Structure of the LC cell with hole diameter of 7mm. Reproduced from [Ch2 65], fig. 1, p. L571.

Figure. 3.3. Interference pattern of the hole etched region obtained using polarized light. The “applied voltage” is 52V AC. Reproduced from [Ch2 65], fig. 4, L572.

The same group reports another design for large hole sizes which create an inhomogeneous electric field [10]. In this design, the cell is designed to gradually destroy the inhomogeneous electric field, created by the “applied voltage”, by applying increasing voltages at the center of the hole region using an additional electrode. This voltage we define as the “additional voltage”. The images of the hole region of the cell, without any application of
voltage at the center or with the application of small voltages have been presented in this thesis as a review of what has been reported in the literature.

Two different hole sizes can be looked into in this design. Corresponding to each hole size, the design of the LC cell, the experimental setup to characterize the samples and the image of the hole region have been provided.

**Hole diameter:** 4.3 mm

**Design:** Fig. 4.4 shows the schematic of the cell design. The upper ITO is separated from the Al electrode by a 70μm glass slab. The thickness of the glass slab which is coated with Al is 1.3 mm. The thickness of the LC layer is 130 μm. The LC used has Δε = 16.8 and has a pretilt angle = 2.5°.

![Diagram of LC cell](image)

Figure. 4.4. Structure of the LC cell with hole diameter of 4.3mm and having three electrode layers. Reproduced from [10], fig. 1, p. 712.

**Experimental setup:** Fig. 4.5 shows the schematic of the experimental setup. A 633nm He-Ne laser is used. The LC cell is kept before a polarizer and a CCD camera is placed before the focus of the laser light. The second analyzer is not being used here. A function generator provides the voltage to the cell [10].
**Image:** The image of the hole etched region is obtained with the cell placed before polarized light. The “applied voltage” is 70V AC and the “additional voltage” is 5V AC. This is shown in fig. 4.6.

Figure. 4.5. Experimental set-up. Reproduced from [10], fig. 6, p. 716.

![Experimental set-up diagram](image)

Figure. 4.6. Interference pattern of the hole etched region obtained using polarized light. The “applied voltage” is 70V and the “additional voltage” is 5V. Reproduced from [10], fig. 7, p. 717.

The second design fig. 4.7 is similar to the first design except that the hole size is different. The experimental setup also uses crossed polarizers instead of just one polarizer [6]
**Hole diameter:** 4.5 mm

**Design:** Fig. 4.7 shows the schematic of the cell design. The upper ITO is separated from the Al electrode by a 70μm glass slab. The thickness of the glass slab which is coated with Al is 1.3 mm. The thickness of the LC layer is 130 μm. The LC used has $\Delta \varepsilon = 16.8$ and has a pretilt angle = $2.5^\circ$.

**Experimental setup:** Fig. 4.8 shows the schematic of the experimental setup. A 633nm He-Ne laser is used for characterization. The LC cell is kept between a polarizer and an analyzer which are at right angles with each other. The rubbing direction of the cell is at $45^\circ$ to each of the polarizer. A function generator provides the voltage to the cell.

**Image:** The image of the hole etched region is obtained with the cell placed between crossed polarizers. The “applied voltage” is 70V AC and no “additional voltage” is applied. This is shown in fig. 4.9.
Figure 4.7. Structure of the LC cell having hole diameter of 4.5mm and having three electrode layers. Reproduced from [6], fig. 1, p. 6408.

![Experimental set-up](image)

Figure 4.8. Experimental set-up. Reproduced from [6], fig. 4, p. 6409.

![Interference pattern](image)

Figure 4.9. Interference pattern of the hole etched region obtained using polarized light. The “applied voltage” is 70V and no “additional voltage” is applied. Reproduced from [6], fig. 9, p. 6411.

In all these above designs the inhomogeneous electric field creates an interference pattern, which goes all the way to the center of the hole region. This has been shown previously in fig. 4.3, 4.6
and 4.9. It is thus possible to obtain an inhomogeneous electric field for holes with large diameter of upto 7mm in hole diameter. To explain the interference pattern we discuss the interaction of the LC material with the polarized light.

4.4 Analysis of LC samples placed between crossed polarizers

- Interaction of the LC material with polarized light

Liquid crystal acts as a phase retarder. Hence the interaction of LC with polarized light is analogous to that between a phase retarder and polarized light [12]. The setup of the sample between two crossed polarizers is shown in fig. 4.10 and the electric field components, with a LC molecule between two crossed polarizers is shown in fig. 4.11.

Figure. 4.10. The LC sample placed between two crossed polarizers. The rubbing direction has been shown in the sample with an arrow [12].
The angle that the LC director $\hat{n}$ makes with the first polarizer is denoted by $\alpha$.

Figure. 4.11. The orientation of the LC molecule and the electric field components, with the cell placed between the polarizer and the analyzer which are perpendicular to each other. The figure developed from the Liquid Crystal Institute coursework.

Light enters through the polarizer and come out of the analyzer. From fig. 4.11, resolving the components, we get $E_{PY}$ and $E_{PX}$.

$$E_{PY} = E_P \cos(\alpha) \quad (4.4.1)$$

$$E_{PX} = E_P \sin(\alpha) \quad (4.4.2)$$
The LC material acts as a phase retarder. The phase retarder provides a phase shift given by \( \exp(i\varphi_y) \) along \( E_{PY} \) and \( \exp(i\varphi_x) \) along \( E_{PX} \).

Consequently, we can obtain \( E_{PYA} \) and \( E_{PXA} \).

\[
E_{PYA} = E_p \cos(\alpha) \sin(\alpha) \exp(i\varphi_y) \quad (4.4.3)
\]

\[
E_{PXA} = E_p \sin(\alpha) \cos(\alpha) \exp(i\varphi_x) \quad (4.4.4)
\]

The field along the analyzer is given as a sum of \( E_{PYA} \) and \( E_{PXA} \). This is given as \( E_A \). \( E_{PYA} \) and \( E_{PXA} \) being vectors opposite to each other, we can obtain eq. 4.4.5 and 4.4.6

\[
E_A = E_p \cos(\alpha) \sin(\alpha) \left\{ \exp(i\varphi_y) - \exp(i\varphi_x) \right\} \quad (4.4.5)
\]

\[
E_A = \frac{1}{2} E_p \sin(2\alpha) \left\{ \exp(i\varphi_y) - \exp(i\varphi_x) \right\} \quad (4.4.6)
\]

The intensity of light reaching the analyzer is square of the amplitude of the electric field along the analyzer.

The amplitude along the analyzer is given by \( |E_A| \) and hence we obtain eq. 4.4.7.
\[ I = |E_A|^2 = I_0 \sin^2(2\alpha)\sin^2 \left( \frac{\Delta \varphi}{2} \right) \]  

(4.4.7)

where, \( I \) is the intensity of light coming out of the analyzer, \( I_0 = |E_P|^2 \) and the phase shift \( \Delta \varphi = \varphi_y - \varphi_x \). This intensity of light is maximized when the LC molecule is at 45° to the polarizer. The intensity is then given as in eq. 4.4.8.

\[ I = I_0 \sin^2 \left( \frac{\Delta \varphi}{2} \right) \]  

(4.4.8)

As seen from eq. 4.4.8, when the phase shift \( \Delta \varphi \) is equal to 2\( \pi \), the intensity is equal to zero and it appears dark. The intensity is brightest when the phase shift is equal to \( \pi \).

Eq. 4.4.8 can be used to study the interference pattern produced inside the hole etched electrode region of a hole patterned LC cell, placed between two crossed polarizers. The interference pattern can be measured by plotting the corresponding phase profile. The interference pattern and the plot is different for cells with planar and homeotropic alignment. Let’s first consider the case of planar alignment. Two different schematics of hole regions with planar alignment has been shown in fig. 4.12(a) and 4.13(a). The cell is at 45° to the polarizer. In the first case, as in fig. 4.13(a), there is a large region at the center where the re-orientation of the LC molecules have not occurred. Under crossed polarizers this region appears bright. The interference rings are thus seen
for a limited region within the hole. In the second case as in fig. 4.13(a), the interference rings extend all the way to the center starting from the edge. This schematic is similar to fig. 4.6. The corresponding phase profiles are shown in fig. 4.12(b) and 4.13(b). Each dark ring represents a phase profile of $2\pi$.

Figure. 4.12. (a) Schematics of the inside of a hole etched electrode region placed between crossed polarizers, of a hole patterned LC cell with planar alignment and with the application of voltage (b) corresponding phase profile obtained from the interference pattern. The voltage is not high enough to re-orient the LC molecules at the center of the hole region. The interference rings are thus seen for a limited region within the hole.
Figure 4.13. (a) Schematics of the inside of a hole etched electrode region placed between crossed polarizers, of a hole patterned LC cell with planar alignment and with the application of voltage (b) corresponding phase profile obtained from the interference pattern. The voltage is enough to re-orient the LC molecules at the center of the hole region. The interference rings extend all the way to the center starting from the edge.

For LC cells with homeotropic alignment, the schematic of the hole etched region under crossed polarizers is as shown in fig. 4.14(a). The electric field is not high enough to reorient the LC molecules at the very center of the cell which under crossed polarizers appear dark. In the
present example, the electric field does not re-orient a large portion of the hole etched region. The interference rings are seen only in the region \( r - r_0 \). The corresponding phase profile is shown in fig. 4.14(b). This cell design is the same as we use in our experiments.

Figure. 4.14. (a) Schematics of the inside of a hole etched electrode region placed between crossed polarizers, of a hole patterned LC cell with homeotropic alignment and with the application of voltage (b) corresponding phase profile obtained from the interference pattern. The voltage is not high enough to re-orient the LC molecules at the center of the hole region.
4.5 Results of hole patterned cell with a 1 mm circular hole etched region with a PI using an initial PI:Solvent ratio of 1:1

4.5.1 Results from outside the hole edged region

A hole patterned LC cell with a small etched region and with the radius of the etched region being 1 mm has been characterized. Both AC as well as DC voltages has been applied in order to compare the electro-optic response. Outside the etched region, the electro-optic response is as shown in fig. 4.15. The cell thickness is ~ 50 μm. The cell has been prepared with PI:Solvent ratio of 1:1 resulting in a PI thickness of ~0.75 μm (fig. 3.5). Initially the homeotropic orientation ensures that the cell appears dark between crossed polarizers. At 2V AC the outside of the hole region appears bright. This can be compared with a Fréedericksz distortion voltage of 1.5V as estimated previously for our cell and 3V as reported [13].
Figure. 4.15. Crossed polarized images of hole patterned LC cells having an etched region of hole diameter 1 mm. AC as well as DC voltages have been applied to the cell (a) voltage OFF (b) 2V AC (c) 2V DC (d) 6V DC. The PI:Solvent ratio used in the fabrication of the cell is 1:1. These images are from outside the hole etched region.

When 2V DC is applied to the cell, the re-orientation does not occur and with the cell kept between crossed polarizers it still appears dark. At 6V DC, the entire region, outside of the hole etched electrode, appears completely bright. This can be compared to values reported before. The DC voltage at which re-orientation occurs have been reported to be ~5V for a 25 μm thick cell [13]. For an impurity content of $10^{-9}$ ions/ molecule, the DC voltage at which re-orientation occurs have been reported to be 1V for a 10 μm thick cell [14].
4.5.2 Results from inside the hole edged region

Inside the electrode etched region, the electro-optic response is as shown in fig. 4.16.

Figure. 4.16. Crossed polarized images of hole patterned LC cells showing the evolution of the electro-optic response inside the cell. The cell has an etched region of radius 1 mm and AC as well as DC voltages have been applied to the cell. (a) voltage off (b) 16V AC (c) 18V AC (d) 16V DC (e) 18V DC. The PI:Solvent ratio used in the fabrication of the cell is 1:1. The pictures have been taken 5 minutes after switching on the voltage.

Without application of voltage, the initial LC orientation being homeotropic, the cell appears dark under crossed polarizers. This is shown in fig. 4.15 (a). When the voltage is switched on, with the application of AC voltage of 2V, the outside of the cell appears bright (fig. 4.15(b)).
In the inside of the cell, an inhomogeneous electric field is created. At 2V AC, inside of the hole region appears dark, which means that the electric field fails to re-orient the LC molecules inside the hole region of the cell. Much above the Freedrickz distortion voltage the reorientation of the LC molecules within the etched region gets complete at 16V AC (fig. 4.16(b)). This can be compared to that reported in the literature of 14V AC for a cell of $\Delta \varepsilon = -0.43$, $\Delta n = 0.25$ and thickness of the cell = 50 $\mu$m [Ch3 4]. The comparison is shown in fig. 4.17 (a) and (b). The corresponding parameters for our cell is $\Delta \varepsilon = -5$, $\Delta n = 0.15$ and thickness of the cell ~ 50 $\mu$m.

Figure. 4.17. (a) Crossed polarized image of the etched region of a hole patterned LC cell with 600$\mu$m hole diameter. The $\Delta \varepsilon = -0.43$, $\Delta n = 0.25$, thickness of the cell = 50 $\mu$m and applied voltage of 14V AC. Reproduced from [Ch3 4], fig. 2, p. 683 (b) crossed polarized image of the cell used in the experiments with the etched region having a hole diameter of 1 mm. The $\Delta \varepsilon = -5$, $\Delta n = 0.15$, thickness of the cell ~ 50 $\mu$m and applied voltage of 16V AC. For DC voltages, at 2V DC, we do not see any reorientation both inside as well as outside the cell (fig. 4.15(c)). This is indicated by the dark region with the cell placed between crossed polarizers, where the Fréedericksz distortion voltage of 1.5V has not been reached. When the voltage is increased to 16V DC, the voltage at which the
AC voltage completely reorients the inside of the electrode etched region, a dark region within the hole etched region is still present (fig. 4.16(d)). However for this voltage, large parts within the hole etched region gets re-oriented as they appear bright with the cell kept between crossed polarizers. The voltage at which the re-orientation of the LC molecules get completed is further at 18V DC (fig. 4.16(e)).

In order to measure the electro-optic response, the phase retardation profiles for AC and DC voltages are measured using the corresponding crossed polarized images. While discussing the phase retardation profiles, it is important to define three terms: “completion”, “no-completion” and “penetration”. “Completion” refers to the completion of the re-orientation of the LC molecules inside the hole etched region. This is as shown in fig. 4.16(c) for AC voltage and fig. 4.16 (e) for DC voltage. Thus “no completion” is defined as the presence of a significantly large dark region inside the hole etched region, as seen with the cell kept between crossed polarizers, even though the reorientation of the LC molecules have already occurred for some portions of the hole region. This is as shown in fig. 4.16(d). “Penetration” refers to the texture of the LC cell kept between crossed polarizers where planarization of certain portion of the hole region have occurred. Consequently the corresponding phase retardation profile, for inside the hole, does not go all the way to the edge but starts outside the region of planarization.

A comparison of the phase retardation profiles between the AC and the DC voltages is shown below. The phase retardation profile, for AC voltages, is shown in fig. 4.18. As can be seen from the phase retardation profile it goes all the way to the edge. The DC phase profiles on the other hand have a different shape as compared to AC phase profiles. This is shown in fig. 4.19. There is “penetration” of the phase profiles in case of DC voltage which is strongly visible for 16V and 18V. This is not present in the phase profiles obtained for the AC voltages.
Figure 4.18. Phase retardation profiles obtained inside the etched region using AC voltages for the hole patterned cell. The etched region of the cell has a diameter equal to 1 mm. The phase retardation profile is measured along the diameter. The PI:Solvent ratio used in the fabrication of the cell is 1:1.
Figure 4.19. Phase retardation profiles obtained inside the etched region using DC voltages for the hole patterned cell. The etched region of the cell has a diameter equal to 1 mm. The phase retardation profile is measured along the diameter. The PI:Solvent ratio used in the fabrication of the cell is 1:1.

A comparison of phase profiles using 16V AC and 18V DC, the voltages at which there is “completion” in the reorientation using AC and DC voltages respectively, is shown in fig. 4.20. There are two important observations. The DC phase profile shows “penetration” at 16V and yet there is “no completion” of the re-orientation of the LC molecules inside the etched region. This is shown in fig. 4.16(d). There is a dark region, which shows that the Fréedericksz distortion voltage has not been reached in that region. Using AC voltage there is no “penetration” although there is “completion” of the re-orientation of the LC molecules inside the etched region at 16V.
Figure 4.20. Comparison of the phase retardation profiles obtained inside the etched region of a hole patterned cell, at the “completion voltage”, using AC and DC. The etched region of the cell has a diameter equal to 1 mm. The phase retardation profile is measured along the diameter. The PI:Solvent ratio used in the fabrication of the cell is 1:1.

In the following sections, the voltages at which the re-orientation occurs outside the hole-etched region for AC and DC voltages, the “completion voltage” inside the hole etched region using both AC as well as DC voltages and the corresponding phase profiles have been provided for samples with different PI thicknesses. The initial PI:Solvent ratio determines the PI thicknesses.
4.6 Results of hole patterned cell with a 1 mm circular hole etched region with a PI using an initial PI:Solvent ratio of 1:2

4.6.1 Results from outside the hole edged region

Figure 4.21. Crossed polarized images of hole patterned LC cells having an etched region of hole diameter 1 mm. AC as well as DC voltages have been applied to the cell (a) voltage OFF (b) 2V AC (c) 2V DC (d) 5.5V DC. The PI:Solvent ratio used in the fabrication of the cell is 1:2. These images are from outside the hole etched region.

Without application of voltage, the initial LC orientation being homeotropic, the cell appears dark under crossed polarizers. This is shown in fig. 4.21 (a). When the voltage is switched on, with the application of AC voltage of 2V, the outside of the cell appears bright (fig. 4.21(b)). For DC voltages, at 2V DC, we do not see any reorientation both inside as well as outside the cell (fig. 64)
4.21(c)). This is indicated by the dark region with the cell placed between 4.21(c)). This is indicated by the dark region with the cell placed between crossed polarizers. Outside the hole etched region the reorientation occurs at 5.5V. This is shown in fig 4.21(d).

4.6.2 Results from inside the hole edged region

Inside the electrode etched region, the electro-optic response is as shown in fig. 4.22.

Figure. 4.22. Crossed polarized images of hole patterned LC cells showing the evolution of the electro-optic response inside the cell. The cell has an etched region of diameter 1 mm and AC as well as DC voltages have been applied to the cell. (a) voltage off (b) 15.5V AC (c) 22V AC (d) 15.5V DC (e) 22V DC. The PI:Solvent ratio used in the fabrication of the cell is 1:2. The pictures have been taken 5 minutes after switching on the voltage.
Inside the hole region when the voltage is increased to 15.5V DC, the voltage at which the AC voltage completely reorients the inside of the electrode etched region, a dark region within the hole etched region is still present (fig. 4.15(d)). The “completion voltage” for DC is 22V DC (fig. 4.22(e)).

Figure. 4.23. Phase retardation profiles obtained inside the etched region using AC voltages for the hole patterned cell. The etched region of the cell has a diameter equal to 1 mm. The phase retardation profile is measured along the diameter. The PI:Solvent ratio used in the fabrication of the cell is 1:2.
Figure 4.24. Phase retardation profiles obtained inside the etched region using DC voltages for the hole patterned cell. The etched region of the cell has a diameter equal to 1 mm. The phase retardation profile is measured along the diameter. The PI:Solvent ratio used in the fabrication of the cell is 1:2.
Figure. 4.25. Comparison of the phase retardation profiles obtained inside the etched region of a hole patterned cell, at the “completion voltage”, using AC and DC. The etched region of the cell has a diameter equal to 1 mm. The phase retardation profile is measured along the diameter. The PI:Solvent ratio used in the fabrication of the cell is 1:2.

The phase retardation profile, for AC voltages, is shown in fig. 4.23. As can be seen from the phase retardation profile it goes all the way to the edge. The DC phase profiles on the other hand have a different shape as compared to AC phase profiles. This is shown in fig. 4.24. There is “penetration” of the phase profiles in case of DC voltage which is strongly visible for 15.5V and 22V and which is not present in the phase profiles obtained for the AC voltages.

A comparison of phase profiles using 15.5V AC and 22V DC, the voltages at which there is “completion” in the reorientation using AC and DC voltages respectively, is shown in fig. 4.25. There are two important observations.
The DC phase profile shows "penetration" at 22V which is the completion voltage. Using AC voltage there is no "penetration" although there is "completion" of the re-orientation of the LC molecules inside the etched region at 15.5V.

4.7 Results of hole patterned cell with a 1 mm circular hole etched region with a PI using an initial PI:Solvent ratio of 1:4

4.7.1 Results from outside the hole edged region

Figure 4.26. Crossed polarized images of hole patterned LC cells having an etched region of hole diameter 1 mm. AC as well as DC voltages have been applied to the cell (a) voltage OFF (b) 2V AC (c) 2V DC (d) 4.5V DC. These images are from outside the hole etched region.

Without application of voltage, the initial LC orientation being homeotropic, the cell appears dark under crossed polarizers. This is shown in fig. 4.26 (a). When the voltage is switched on, with the application of AC voltage of 2V, the outside of the cell appears bright (fig. 4.26(b)).
For DC voltages, at 2V DC, we do not see any reorientation both inside as well as outside the cell (fig. 4.26(c)). This is indicated by the dark region with the cell placed between crossed polarizers. Outside the hole etched region the reorientation occurs at 4.5V. This is shown in fig 4.26(d).

### 4.7.2 Results from inside the hole edged region

Inside the electrode etched region, the electro-optic response is as shown in fig. 4.27.

Figure. 4.27. Crossed polarized images of hole patterned LC cells showing the evolution of the electro-optic response inside the cell. The cell has an etched region of diameter 1 mm and AC as
well as DC voltages have been applied to the cell. (a) voltage off (b) 15V AC (c) 23V AC (d) 15V DC (e) 23V DC. The PI:Solvent ratio used in the fabrication of the cell is 1:4. The pictures have been taken 5 minutes after switching on the voltage.

Inside the hole region when the voltage is increased to 15V DC, the voltage at which the AC voltage completely reorients the inside of the electrode etched region, a dark region within the hole etched region is still present (fig. 4.27(d)). The “completion voltage” is at 23V DC (fig. 4.27(e)).

Figure. 4.28. Phase retardation profiles obtained inside the etched region using AC voltages for the hole patterned cell. The etched region of the cell has a diameter equal to 1 mm. The phase retardation profile is measured along the diameter. The PI:Solvent ratio used in the fabrication of the cell is 1:8.
Figure 4.29. Phase retardation profiles obtained inside the etched region using DC voltages for the hole patterned cell. The etched region of the cell has a diameter equal to 1 mm. The phase retardation profile is measured along the diameter. The PI:Solvent ratio used in the fabrication of the cell is 1:8.
Figure 4.30. Comparison of the phase retardation profiles obtained inside the etched region of a hole patterned cell, at the “completion voltage”, using AC and DC. The etched region of the cell has a diameter equal to 1 mm. The phase retardation profile is measured along the diameter. The PI:Solvent ratio used in the fabrication of the cell is 1:4.

The phase retardation profile, for AC voltages, is shown in fig. 4.28. As can be seen from the phase retardation profile it goes all the way to the edge. The DC phase profiles on the other hand have a different shape as compared to AC phase profiles. This is shown in fig. 4.29. There is “penetration” of the phase profiles in case of DC voltage which is strongly visible for 15V and 23V and which is not present in the phase profiles obtained for the AC voltages. A comparison of phase profiles using 15V AC and 23V DC, the voltages at which there is “completion” in the reorientation using AC and DC voltages respectively, is shown in fig. 4.30. There are two important observations.
The DC phase profile shows “penetration” at 23V. Using AC voltage there is no “penetration” although there is “completion” of the re-orientation of the LC molecules inside the etched region at 15V.

4.8 Results of hole patterned cell with a 1 mm circular hole etched region with a PI using an initial PI:Solvent ratio of 1:8

4.8.1 Results from outside the hole edged region

Figure 4.31. Crossed polarized images of hole patterned LC cells having an etched region of hole diameter 1 mm. AC as well as DC voltages have been applied to the cell (a) voltage OFF (b) 2V AC (c) 2V DC (d) 2.5V DC. These images are from outside the hole etched region. Without application of voltage, the initial LC orientation being homeotropic, the cell appears dark under
crossed polarizers. This is shown in fig. 4.31 (a). When the voltage is switched on, with the application of AC voltage of 2V, the outside of the cell appears bright (fig. 4.31(b)). For DC voltages, at 2V DC, we do not see any reorientation both inside as well as outside the cell (fig. 4.21(c)). This is indicated by the dark region with the cell placed between crossed polarizers. Outside the hole etched region the reorientation occurs at 2.5V. This is shown in fig. 4.31(d).

4.8.2 Results from inside the hole edged region

Inside the electrode etched region, the electro-optic response is as shown in fig. 3.32.

Figure. 4.32. Crossed polarized images of hole patterned LC cells showing the evolution of the electro-optic response inside the cell. The cell has an etched region of diameter 1 mm and AC as
well as DC voltages have been applied to the cell. (a) voltage off (b) 15V AC (c) 24V AC (d) 15V DC (e) 24V DC. The PI:Solvent ratio used in the fabrication of the cell is 1:8. The pictures have been taken 5 minutes after switching on the voltage.

Inside the hole region when the voltage is increased to 15V DC, the voltage at which the AC voltage completely reorients the inside of the electrode etched region, a dark region within the hole etched region is still present (fig. 4.32(d)). The “completion voltage” is at 24V DC (fig. 4.32(e)).

![Figure. 4.33. Phase retardation profiles obtained inside the etched region using AC voltages for the hole patterned cell. The etched region of the cell has a diameter equal to 1 mm. The phase retardation profile is measured along the diameter. The PI:Solvent ratio used in the fabrication of the cell is 1:8.](image-url)
Figure 4.34. Phase retardation profiles obtained inside the etched region using DC voltages for the hole patterned cell. The etched region of the cell has a diameter equal to 1 mm. The phase retardation profile is measured along the diameter. The PI:Solvent ratio used in the fabrication of the cell is 1:8.
Figure 4.35. Comparison of the phase retardation profiles obtained inside the etched region of a hole patterned cell, at the “completion voltage”, using AC and DC. The etched region of the cell has a diameter equal to 1 mm. The phase retardation profile is measured along the diameter. The PI:Solvent ratio used in the fabrication of the cell is 1:8.

The phase retardation profile, for AC voltages, is shown in fig. 4.33. As can be seen from the phase retardation profile it goes all the way to the edge. The DC phase profiles on the other hand have a different shape as compared to AC phase profiles. This is shown in fig. 4.34. There is “penetration” of the phase profiles in case of DC voltage which is strongly visible for 15V and 24V and which is not present in the phase profiles obtained for the AC voltages. A comparison of phase profiles using 15V AC and 24V DC, the voltages at which there is “completion” in the reorientation using AC and DC voltages respectively, is shown in fig. 4.35. There are two important observations. The DC phase profile shows “penetration” at 15V and yet
there is “no completion” of the re-orientation of the LC molecules inside the etched region. This is shown in fig. 4.32(d). Using AC voltage there is no “penetration” although there is “completion” of the re-orientation of the LC molecules inside the etched region at 15V.

4.9 Results of hole patterned cell with a 1 mm circular hole etched region with a PI using an initial PI:Solvent ratio of 1:16

4.9.1 Results from outside the hole edged region

Figure. 4.36. Crossed polarized images of hole patterned LC cells having an etched region of hole diameter 1 mm. AC as well as DC voltages have been applied to the cell (a) voltage OFF (b) 2V AC (c) 2V DC (d) 5.5V DC. These images are from outside the hole etched region.
Without application of voltage, the initial LC orientation being homeotropic, the cell appears dark under crossed polarizers. This is shown in fig. 4.36 (a). When the voltage is switched on, with the application of AC voltage of 2V, the outside of the cell appears bright (fig. 4.36(b)).

### 4.9.2 Results from inside the hole edged region

Inside the electrode etched region, the electro-optic response is as shown in fig. 4.37.

![Crossed polarized images of hole patterned LC cells showing the evolution of the electro-optic response inside the cell. The cell has an etched region of diameter 1 mm and AC as](image)

Figure. 4.37. Crossed polarized images of hole patterned LC cells showing the evolution of the electro-optic response inside the cell. The cell has an etched region of diameter 1 mm and AC as
well as DC voltages have been applied to the cell. (a) voltage off (b) 17V AC (c) 28V AC (d) 17V DC (e) 28V DC. The PI:Solvent ratio used in the fabrication of the cell is 1:16. The pictures have been taken 5 minutes after switching on the voltage.

For DC voltages, at 2V DC, we do not see any reorientation both inside as well as outside the cell (fig. 4.36(c)). This is indicated by the dark region with the cell placed between crossed polarizers. Outside the hole etched region the reorientation occurs at 5.5V. This is shown in fig. 4.36(d).

Inside the hole region when the voltage is increased to 17V DC, the voltage at which the AC voltage completely reorients the inside of the electrode etched region, a dark region within the hole etched region is still present (fig. 4.37(d)). The “completion voltage” is at 28V DC (fig. 4.37(e)).

![Figure 4.38](image)

Figure 4.38. Phase retardation profiles obtained inside the etched region using AC voltages for the hole patterned cell. The etched region of the cell has a diameter equal to 1 mm. The phase
retardation profile is measured along the diameter. The PI:Solvent ratio used in the fabrication of the cell is 1:16.

Figure 4.39. Phase retardation profiles obtained inside the etched region using DC voltages for the hole patterned cell. The etched region of the cell has a diameter equal to 1 mm. The phase retardation profile is measured along the diameter. The PI:Solvent ratio used in the fabrication of the cell is 1:16.
Figure 4.40. Comparison of the phase retardation profiles obtained inside the etched region of a hole patterned cell, at the “completion voltage”, using AC and DC. The etched region of the cell has a diameter equal to 1 mm. The phase retardation profile is measured along the diameter. The PI:Solvent ratio used in the fabrication of the cell is 1:16.

The phase retardation profile, for AC voltages, is shown in fig. 4.38. As can be seen from the phase retardation profile it goes all the way to the edge. The DC phase profiles on the other hand have a different shape as compared to AC phase profiles. This is shown in fig. 4.39. There is “penetration” of the phase profiles in case of DC voltage which is strongly visible for 17V and 28V and which is not present in the phase profiles obtained for the AC voltages. A comparison of phase profiles using 17V AC and 28V DC, the voltages at which there is “completion” in the reorientation using AC and DC voltages respectively, is shown in fig. 4.40. There are two important observations. The DC phase profile shows “penetration” at 17V and yet
there is “no completion” of the re-orientation of the LC molecules inside the etched region. This is shown in fig. 4.37(d). Using AC voltage there is no “penetration” although there is “completion” of the re-orientation of the LC molecules inside the etched region at 17V.

4.10 Results of hole patterned cell with a 2 mm circular hole etched region with a PI using an initial PI:Solvent ratio of 1:1

Inside the electrode etched region, the electro-optic response is as shown in fig. 4.41.

Figure. 4.41. Crossed polarized images of hole patterned LC cells showing the evolution of the electro-optic response inside the cell. The cell has an etched region of diameter 2 mm and AC as well as DC voltages have been applied to the cell. (a) voltage off (b) 34V AC (c) 38V AC (d) 34V DC (e) 38V DC. The PI:Solvent ratio used in the fabrication of the cell is 1:1. The pictures have been taken 5 minutes after switching on the voltage.
Inside the hole region the “completion voltage” is 34V AC. Using a similar DC voltage, the reorientation is “not complete”. There is a dark region within the hole etched region which is still present (fig. 4.41(d)). The “completion voltage” is at 38V DC (fig. 4.41(e)).

Figure. 4.42. Phase retardation profiles obtained inside the etched region using AC voltages for the hole patterned cell. The etched region of the cell has a diameter equal to 2 mm. The phase retardation profile is measured along the diameter. The PI:Solvent ratio used in the fabrication of the cell is 1:1.
Figure 4.43. Phase retardation profiles obtained inside the etched region using DC voltages for the hole patterned cell. The etched region of the cell has a diameter equal to 2 mm. The phase retardation profile is measured along the diameter. The PI:Solvent ratio used in the fabrication of the cell is 1:1.
Figure 4.44. Comparison of the phase retardation profiles obtained inside the etched region of a hole patterned cell, at the “completion voltage”, using AC and DC. The etched region of the cell has a diameter equal to 2 mm. The phase retardation profile is measured along the diameter. The PI:Solvent ratio used in the fabrication of the cell is 1:1.

The phase retardation profile, for AC voltages, is shown in fig. 4.42. As can be seen from the phase retardation profile it goes all the way to the edge. The DC phase profiles on the other hand have a different shape as compared to AC phase profiles. This is shown in fig. 4.43. There is “penetration” of the phase profiles in case of DC voltage which is strongly visible for 34V and 38V and which is not present in the phase profiles obtained for the AC voltages. A comparison of phase profiles using 34V AC and 38V DC, the voltages at which there is “completion” in the reorientation using AC and DC voltages respectively, is shown in fig. 4.44. There are two
important observations. The DC phase profile shows “penetration” at 34V and yet there is “no completion” of the re-orientation of the LC molecules inside the etched region. This is shown in fig. 4.41(d). Using AC voltage there is no “penetration” although there is “completion” of the re-orientation of the LC molecules inside the etched region at 34V.

4.11 Results of hole patterned cell with a 2 mm circular hole etched region with a PI using an initial PI:Solvent ratio of 1:2

Inside the electrode etched region, the electro-optic response is as shown in fig. 4.45.

Figure. 4.45. Crossed polarized images of hole patterned LC cells showing the evolution of the electro-optic response inside the cell. The cell has an etched region of diameter 2 mm and AC as well as DC voltages have been applied to the cell. (a) voltage off (b) 33V AC (c) 46V AC (d) 33V
DC (e) 46V DC. The PI:Solvent ratio used in the fabrication of the cell is 1:2. The pictures have been taken 5 minutes after switching on the voltage.

Inside the hole region the “completion voltage” is 33V AC. Using a similar DC voltage, the reorientation is “not complete”. There is a dark region within the hole etched region which is still present (fig. 4.45(d)). The “completion voltage” is at 46V DC (fig. 4.45(e)).

Figure. 4.46. Phase retardation profiles obtained inside the etched region using AC voltages for the hole patterned cell. The etched region of the cell has a diameter equal to 2 mm. The phase retardation profile is measured along the diameter. The PI:Solvent ratio used in the fabrication of the cell is 1:2.
Figure 4.47. Phase retardation profiles obtained inside the etched region using DC voltages for the hole patterned cell. The etched region of the cell has a diameter equal to 2 mm. The phase retardation profile is measured along the diameter. The PI:Solvent ratio used in the fabrication of the cell is 1:2.
Figure 4.48. Comparison of the phase retardation profiles obtained inside the etched region of a hole patterned cell, at the “completion voltage”, using AC and DC. The etched region of the cell has a diameter equal to 2 mm. The phase retardation profile is measured along the diameter. The PI:Solvent ratio used in the fabrication of the cell is 1:2.

The phase retardation profile, for AC voltages, is shown in fig. 4.46. As can be seen from the phase retardation profile it goes all the way to the edge. The DC phase profiles on the other hand have a different shape as compared to AC phase profiles. The DC phase retardation profiles are shown in fig. 4.47. There is “penetration” of the phase profiles in case of DC voltage which is strongly visible for 33V and 46V and which is not present in the phase profiles obtained for the AC voltage.
A comparison of phase profiles using 33V AC and 46V DC, the voltages at which there is “completion” in the reorientation using AC and DC voltages respectively, is shown in fig. 4.48. There are two important observations. The DC phase profile shows “penetration” at 33V and yet there is “no completion” of the re-orientation of the LC molecules inside the etched region. This is shown in fig. 4.45(d). Using AC voltage there is no “penetration” although there is “completion” of the re-orientation of the LC molecules inside the etched region at 33V.

4.12 Results of hole patterned cell with a 2 mm circular hole etched region with a PI using an initial PI:Solvent ratio of 1:4

Inside the electrode etched region, the electro-optic response is as shown in fig. 4.49.

Figure. 4.49. Crossed polarized images of hole patterned LC cells showing the evolution of the electro-optic response inside the cell. The cell has an etched region of diameter 2 mm and AC as
well as DC voltages have been applied to the cell. (a) voltage off (b) 33V AC (c) 50V AC (d) 33V DC (e) 50V DC. The PI:Solvent ratio used in the fabrication of the cell is 1:4. The pictures have been taken 5 minutes after switching on the voltage.

Inside the hole region the “completion voltage” is 33V AC. Using a similar DC voltage, the reorientation is “not complete”. There is a dark region within the hole etched region which is still present (fig. 4.49(d)). The “completion voltage” is at 50V DC (fig. 4.49(e)).

Figure. 4.50. Phase retardation profiles obtained inside the etched region using AC voltages for the hole patterned cell. The etched region of the cell has a diameter equal to 2 mm. The phase retardation profile is measured along the diameter. The PI:Solvent ratio used in the fabrication of the cell is 1:4.
Figure 4.51. Phase retardation profiles obtained inside the etched region using DC voltages for the hole patterned cell. The etched region of the cell has a diameter equal to 2 mm. The phase retardation profile is measured along the diameter. The PI:Solvent ratio used in the fabrication of the cell is 1:4.
Figure. 4.52. Comparison of the phase retardation profiles obtained inside the etched region of a hole patterned cell, at the “completion voltage”, using AC and DC. The etched region of the cell has a diameter equal to 2 mm. The phase retardation profile is measured along the diameter. The PI:Solvent ratio used in the fabrication of the cell is 1:4.

The phase retardation profile, for AC voltages, is shown in fig. 4.50. As can be seen from the phase retardation profile it goes all the way to the edge. The DC phase profiles on the other hand have a different shape as compared to AC phase profiles. This is shown in fig. 4.51. There is “penetration” of the phase profiles in case of DC voltage which is strongly visible for 33V and 50V and which is not present in the phase profiles obtained for the AC voltages. A comparison of phase profiles using 33V AC and 50V DC, the voltages at which there is “completion” in the reorientation using AC and DC voltages respectively, is shown in fig. 4.52. There are two important observations. The DC phase profile shows “penetration” at 33V and yet
there is “no completion” of the re-orientation of the LC molecules inside the etched region. This is shown in fig. 4.49(c). Using AC voltage there is no “penetration” although there is “completion” of the re-orientation of the LC molecules inside the etched region at 33V.

### 4.13 Results of hole patterned cell with a 2 mm circular hole etched region with a PI using an initial PI:Solvent ratio of 1:8

Inside the electrode etched region, the electro-optic response is as shown in fig. 4.53.

![Figure. 4.53. Crossed polarized images of hole patterned LC cells showing the evolution of the electro-optic response inside the cell. The cell has an etched region of diameter 2 mm and AC as](image)

96
well as DC voltages have been applied to the cell. (a) voltage off (b) 33V AC (c) 52V AC (d) 33V DC (e) 52V DC. The PI:Solvent ratio used in the fabrication of the cell is 1:8. The pictures have been taken 5 minutes after switching on the voltage.

Inside the hole region the “completion voltage” is 33V AC. Using a similar DC voltage, the reorientation is “not complete”. There is a dark region within the hole etched region which is still present (fig. 4.53(d)). The “completion voltage” is at 52V DC (fig. 4.53(e)).

Figure. 4.54. Phase retardation profiles obtained inside the etched region using AC voltages for the hole patterned cell. The etched region of the cell has a diameter equal to 2 mm. The phase retardation profile is measured along the diameter. The PI:Solvent ratio used in the fabrication of the cell is 1:8.
Figure 4.55. Phase retardation profiles obtained inside the etched region using DC voltages for the hole patterned cell. The etched region of the cell has a diameter equal to 2 mm. The phase retardation profile is measured along the diameter. The PI:Solvent ratio used in the fabrication of the cell is 1:8.
Figure. 4.56. Comparison of the phase retardation profiles obtained inside the etched region of a hole patterned cell, at the “completion voltage”, using AC and DC. The etched region of the cell has a diameter equal to 2 mm. The phase retardation profile is measured along the diameter. The PI:Solvent ratio used in the fabrication of the cell is 1:8.

The phase retardation profile, for AC voltages, is shown in fig. 4.54. As can be seen from the phase retardation profile it goes all the way to the edge. The DC phase profiles on the other hand have a different shape as compared to AC phase profiles. This is shown in fig. 4.55. There is “penetration” of the phase profiles in case of DC voltage which is strongly visible for 33V and 52V and which is not present in the phase profiles obtained for the AC voltages. A comparison of phase profiles using 33V AC and 52V DC, the voltages at which there is “completion” in the reorientation using AC and DC voltages respectively, is shown in fig. 4.56. There are two important observations. The DC phase profile shows “penetration” at 33V and yet
there is “no completion” of the re-orientation of the LC molecules inside the etched region. This is shown in fig. 4.53(d). Using AC voltage there is no “penetration” although there is “completion” of the re-orientation of the LC molecules inside the etched region at 33V.

### 4.14 Results of hole patterned cell with a 2 mm circular hole etched region with a PI using an initial PI:Solvent ratio of 1:16

Inside the electrode etched region, the electro-optic response is as shown in fig. 4.57.

Figure. 4.57. Crossed polarized images of hole patterned LC cells showing the evolution of the electro-optic response inside the cell. The cell has an etched region of diameter 2 mm and AC as
well as DC voltages have been applied to the cell. (a) voltage off (b) 35V AC (c) 57V AC (d) 35V DC (e) 57V DC. The PI:Solvent ratio used in the fabrication of the cell is 1:16. The pictures have been taken 5 minutes after switching on the voltage.

Inside the hole region the “completion voltage” is 35V AC. Using a similar DC voltage, the reorientation is “not complete”. There is a dark region within the hole etched region which is still present (fig. 4.57(d)). The “completion voltage” is at 57V DC (fig. 4.57(e)).

![Phase retardation profiles](image)

**Figure.** 4.58. Phase retardation profiles obtained inside the etched region using AC voltages for the hole patterned cell. The etched region of the cell has a diameter equal to 2 mm. The phase retardation profile is measured along the diameter. The PI:Solvent ratio used in the fabrication of the cell is 1:16.
Figure. 4.59. Phase retardation profiles obtained inside the etched region using DC voltages for the hole patterned cell. The etched region of the cell has a diameter equal to 2 mm. The phase retardation profile is measured along the diameter. The PI:Solvent ratio used in the fabrication of the cell is 1:16.
Figure. 4.60. Comparison of the phase retardation profiles obtained inside the etched region of a hole patterned cell, at the “completion voltage”, using AC and DC. The etched region of the cell has a diameter equal to 2 mm. The phase retardation profile is measured along the diameter. The PI:Solvent ratio used in the fabrication of the cell is 1:16.

The phase retardation profile, for AC voltages, is shown in fig. 4.58. As can be seen from the phase retardation profile it goes all the way to the edge. The DC phase profiles on the other hand have a different shape as compared to AC phase profiles. This is shown in fig. 4.59. There is “penetration” of the phase profiles in case of DC voltage which is strongly visible for 35V and 57V and which is not present in the phase profiles obtained for the AC voltages.

A comparison of phase profiles using 35V AC and 57V DC, the voltages at which there is “completion” in the reorientation using AC and DC voltages respectively, is shown in fig. 4.60. There are two important observations. The DC phase profile shows “penetration” at 35V and yet
there is “no completion” of the re-orientation of the LC molecules inside the etched region. This is shown in fig. 4.57(d). Using AC voltage there is no “penetration” although there is “completion” of the re-orientation of the LC molecules inside the etched region at 35V.

### 4.15 Results Summary

<table>
<thead>
<tr>
<th>Pl:Solvent</th>
<th>1:1</th>
<th>1:2</th>
<th>1:4</th>
<th>1:8</th>
<th>1:16</th>
</tr>
</thead>
<tbody>
<tr>
<td>Voltage type</td>
<td>AC (V)</td>
<td>DC (V)</td>
<td>AC (V)</td>
<td>DC (V)</td>
<td>AC (V)</td>
</tr>
<tr>
<td>Critical voltage (outside the hole etched region)</td>
<td>2</td>
<td>6</td>
<td>2</td>
<td>5.5</td>
<td>2</td>
</tr>
<tr>
<td>1mm completion</td>
<td>16</td>
<td>18</td>
<td>15.5</td>
<td>22</td>
<td>15</td>
</tr>
<tr>
<td>2mm completion</td>
<td>34</td>
<td>38</td>
<td>33</td>
<td>46</td>
<td>33</td>
</tr>
<tr>
<td>3mm completion</td>
<td>52</td>
<td>59</td>
<td>52</td>
<td>74</td>
<td>54</td>
</tr>
<tr>
<td>4mm completion</td>
<td>97</td>
<td>110</td>
<td>97</td>
<td>136</td>
<td>95.5</td>
</tr>
</tbody>
</table>

Table 1: Critical voltage required to re-orient the LC molecules outside the hole etched region and the voltage corresponding to the completion of the re-orientation in the hole etched region for different hole diameters ranging from 1mm to 4mm.
Figure 4.61. Comparison of the phase retardation profiles obtained inside the etched region of a hole patterned cell, at the “completion voltage”, using DC voltage and for various PI thicknesses. The etched region of the cell has a diameter equal to 2 mm. The phase retardation profile is measured along the diameter.
Figure 4.62. Comparison of the phase retardation profiles obtained inside the etched region of a hole patterned cell, at the minimum “completion voltage” of 38V amongst all samples and using DC voltage. The phase retardation profiles have been plotted at constant voltages and for various PI thicknesses. The etched region of the cell has a diameter equal to 2 mm. The phase retardation profile is measured along the diameter.
4.16 Discussion

Figure 4.63. PI thickness vs. voltage outside the hole etched region

Figure 4.64. PI thickness vs. “completion voltage” for a 1mm hole etched region
In order to drive the liquid crystal cell, AC or DC voltage is applied. This voltage generates an electric field given by [6]

\[ E = -\nabla V \]  

(4.16.1)

where \( E \) is the electric field and \( V \) is the voltage. This field is a spatially distributed one inside the hole patterned electrode region and obeys the Maxwell equation

\[ \nabla \cdot D = 0 \]  

(4.16.2)

where \( D \) is the electric displacement

or \( \nabla \cdot (\varepsilon \cdot E) = 0 \)  

(4.16.3)

where \( \varepsilon \) denotes the dielectric tensor and \( E \) is the electric field.

For the LC material the dielectric tensor is given by
\[(\varepsilon_{LC})_{\alpha\beta} = \varepsilon_{\perp} \delta_{\alpha\beta} + (\varepsilon_{\parallel} - \varepsilon_{\perp}) n_\alpha n_\beta \tag{4.16.4}\]

Where \(\alpha\) and \(\beta\) denote the \(x\), \(y\) and \(z\) components. \(\varepsilon_{\parallel}\) and \(\varepsilon_{\perp}\) denote the parallel and perpendicular components of the dielectric constant with respect to the local LC directors respectively. The director vector is given by \(\hat{n} = (n_x, n_y, n_z)\). The applied electric field creates a local torque on the liquid crystal molecules which is given by \((\varepsilon_{\parallel} - \varepsilon_{\perp})(\hat{n} \cdot \vec{E})(\hat{n} \times \vec{E})/(4 \pi)\) which causes the liquid crystal molecules to be aligned in the direction of the electric field \(\vec{E}\). This torque is countered by the elastic torque of the liquid crystal molecules. The elastic torque is given by

\[K \frac{\partial^2 \theta}{\partial z^2}\]

where \(K\) is the elastic constant and \(\theta\) is the angle between the director \(\hat{n}\) and the unit vector parallel to the \(z\) axis. Equating both the torques, the final equation is given by eq. 4.15.1.5

\[
\frac{(\varepsilon_{\parallel} - \varepsilon_{\perp})(\hat{n} \cdot \vec{E})(\hat{n} \times \vec{E})}{4 \pi} = K \frac{\partial^2 \theta}{\partial z^2} \tag{4.16.5}
\]

This is true both inside as well as outside the hole patterned regions. However inside the hole patterned region there is a spatial distribution of the electric field. This inhomogenous electric field creates a director profile resulting in refractive index profile. This results in corresponding equipotential lines. When voltage is applied to a hole patterned cell with an electrode structure as given in fig. 4.66 it results in equipotential lines as shown in fig. 4.67
Fig. 4.66. Electrode structure of a hole patterned cell with provision to apply two different voltages. Reproduced from [6], fig. 1, p. 6408.

Fig. 4.67. Equipotential lines for the same hole patterned cell after applying two different voltages (a) 70V for Al electrode and 10V for ITO electrode (b) 70V for Al electrode and 20V for ITO electrode. Reproduced from [6], fig. 3, p. 6409.
Two different voltages are applied for the outer ITO electrodes and the inner Al electrode. 70V AC is applied to the outer ITO electrode and a 10 V AC is applied to the Al electrode. That results in equipotential lines as shown in fig. 4.67. A similar set of equipotential lines can be obtained using a cell structure similar to that in my experiments. For such a cell structure the equipotential lines for a half cell are as shown in fig. 4.68. The hole diameter for the simulation in this case is 600 \( \mu m \) and the thickness is 50 \( \mu m \) [Ch3 4].

Fig. 4.68. Equipotential lines for a half cell electrode structure. Reproduced from [Ch3 4], fig. 6, p. 685.

In order to obtain the equipotential lines as given in fig. 4.68, the following conditions need to be applied to solve the Laplace equation using the anisotropic condition. There are three sets of boundary conditions.

For \( r \geq L/2 \), the boundary conditions are the following
\[ u(r,0) = 0 \quad (4.16.6) \]

\[ u(r,d) = U \quad (4.16.7) \]

For \( r < L/2 \), the boundary conditions are the following

\[ u(r,0) = \varphi(r) \quad (4.16.8) \]

\[ E(0,z) = \frac{\partial u(0,z)}{\partial r} = 0 \quad (4.16.9) \]

Parallel components of the electric field, at the boundary for the LC and the glass are the same

\[ E_{\parallel LC} = E_{\parallel gl} \quad (4.16.10) \]

\[ \frac{\partial \varphi(r)_{\parallel LC}}{\partial r} = \frac{\partial \varphi(r)_{\parallel gl}}{\partial r} \quad (4.16.11) \]

Perpendicular components of the electric displacement vectors for the LC and the glass are the same

\[ \varepsilon_{LC} \vec{E}_{nLC} = \varepsilon_{gl} \vec{E}_{ngl} \quad (4.16.12) \]

For \( z < 0 \), the boundary conditions are the following

\[ \varphi(r,-\infty) = 0 \quad (4.16.13) \]

\[ \varphi(\infty,z) = 0 \quad (4.16.14) \]

In Cartesian co-ordinates the Laplace equation in the simplest isotropic form \( \nabla^2 u \) is given by

\[ \nabla^2 u = \frac{\partial^2 u}{\partial x^2} + \frac{\partial^2 u}{\partial y^2} + \frac{\partial^2 u}{\partial z^2} \quad (4.16.15) \]

In cylindrical co-ordinates
\[ \nabla^2 u = \frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial u}{\partial r} \right) + \frac{1}{r^2} \frac{\partial^2 u}{\partial \theta^2} + \frac{\partial^2 u}{\partial z^2} \]  

(4.16.16)

The system being axially symmetric, the cylindrical co-ordinates can be written as

\[ \nabla^2 u = \frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial u}{\partial r} \right) + \frac{\partial^2 u}{\partial z^2} \]  

(4.16.17)

Finite difference method to solve the Laplace equation is given in [15]

Let us consider a region given by rectangle \( L_x, L_y \). We create a mesh on the rectangle. Unit cell of the mesh is a square with \( \Delta \). For each lattice point of the mesh the potential is \( \phi(i,j) \)

Laplace equation is approximated by

\[
\frac{\phi(i + 1,j) + \phi(i - 1,j) - 2 \times \phi(i,j)}{\Delta^2} + \frac{\phi(i,j + 1) + \phi(i,j - 1) - 2 \times \phi(i,j)}{\Delta^2} = 0
\]

i = 0,1,2, ..., \( N_x (N_x = \frac{L_x}{\Delta}) \)

j = 0,1,2, ..., \( N_y (N_y = \frac{L_y}{\Delta}) \)

(4.16.18)

Rearranging equation

\[
\phi(i + 1,j) + \phi(i - 1,j) - 2 \times \phi(i,j) + \phi(i,j + 1) + \phi(i,j - 1) - 2 \times \phi(i,j) = 0
\]

(4.16.19)

\[
\phi(i,j) = \frac{1}{4} \times [\phi(i + 1,j) + \phi(i - 1,j) + \phi(i,j + 1) + \phi(i,j - 1)]
\]

(4.16.20)

From the boundary conditions, the potential at the boundary is given. Let it be \( \phi_0(i,j) \). Using the values of the potential at the nearest neighbor mesh points we can find the potential at any mesh
point. The iterative method of calculation is slow and can be improved by the ‘over relaxation’ method, where

\[ \phi^{r+1}(i,j) = \]

\[ \phi^r(i,j) + \frac{\alpha}{4} [\phi^r(i + 1, j) + \phi^r(i - 1, j) - 2 \phi^r(i, j) + \phi^r(i, j + 1) + \phi^r(i, j - 1) - 2 \phi^r(i, j)] \]  

(4.16.21)

\[ \phi^{r+1}(i,j) = \phi^r(i,j) + \frac{\alpha}{4} \Delta^2 \left[ \frac{\partial^2 \phi^r}{\partial x^2}(i,j) + \frac{\partial^2 \phi^r}{\partial y^2}(i,j) \right] \]  

(4.16.22)

The change \( \Delta \phi^r(i,j) = \phi^{r+1}(i,j) - \phi^r(i,j) \) becomes smaller and smaller

When \( \phi^r(i,j) \) does not change any more

\[ \frac{\partial^2 \phi^r}{\partial x^2}(i,j) + \frac{\partial^2 \phi^r}{\partial y^2}(i,j) = 0 \]  

(4.16.23)

Which means that the Laplace equation is satisfied. The finite difference method, using an over relaxation model, for a 3D anisotropic medium, as in our experiments, in given in [26]

\[ \phi^{r+1}(i,j,k) = \phi^r(i,j,k) + \frac{\alpha}{4} \Delta^2 \left[ \frac{\partial \phi^r}{\partial x}(\varepsilon_{11} \frac{\partial \phi}{\partial x}) + \frac{\partial \phi^r}{\partial y}(\varepsilon_{12} \frac{\partial \phi}{\partial y}) + \frac{\partial \phi^r}{\partial z}(\varepsilon_{13} \frac{\partial \phi}{\partial z}) \right] (i,j,k) \]

(4.16.24)

The mixed derivatives can be written as given in [16]
\[
\left( \frac{\partial^2 \phi}{\partial x \partial y} \right)_{i,j} = \frac{\left( \frac{\partial \phi}{\partial y} \right)_{i+1,j} - \left( \frac{\partial \phi}{\partial y} \right)_{i-1,j}}{2 \Delta x}
\]
\[
\left( \frac{\partial \phi}{\partial y} \right)_{i+1,j} = \frac{\Phi_{i+1,j+1} - \Phi_{i+1,j-1}}{2 \Delta y}
\]
\[
\left( \frac{\partial \phi}{\partial y} \right)_{i-1,j} = \frac{\Phi_{i-1,j+1} - \Phi_{i-1,j-1}}{2 \Delta y}
\]
\[
\left( \frac{\partial^2 \phi}{\partial x \partial y} \right)_{i,j} = \frac{\Phi_{i+1,j+1} - \Phi_{i+1,j-1} - \Phi_{i-1,j+1} + \Phi_{i-1,j-1}}{4 \Delta x \Delta y}
\]

Similarly other mixed value derivatives can be represented as a finite difference method. In order to find out the deformations of the liquid crystal molecules and thereby the tilt angle at each meshpoint, the free energy density has to be considered. The free energy density for the elastic part is given by

\[
f_{\text{elastic}} = \frac{1}{2} K_{11} \left( \nabla \cdot \hat{n} \right)^2 + \frac{1}{2} K_{22} \left( \hat{n} \cdot \nabla \hat{n} \right)^2 + \frac{1}{2} K_{33} \left( \hat{n} \times \nabla \times \hat{n} \right)^2
\]

Where \( K_{11}, K_{22} \) and \( K_{33} \) are splay, twist and bend elastic constants and \( \hat{n} \) is the director.

The director \( \hat{n} \) can be given by \( \hat{n} = n_\rho, n_\varphi, n_z \). For a liquid crystal with homeotropic orientation, the electric field is axially symmetric and thus the twist is missing. \( \theta \) is the angle between z axis and the director \( \hat{n} \).

\[
n_\rho = \sin \theta
\]
\[
n_z = \cos \theta
\]
\[ n_\phi = 0 \tag{4.16.33} \]

\[ \hat{n} = (\sin \theta, 0, \cos \theta) \tag{4.16.34} \]

\[ \nabla \cdot n = -\sin \theta \cdot \theta' \tag{4.16.35} \]

\[ \nabla \cdot n = \cos \theta \cdot \theta' \hat{y} \tag{4.16.36} \]

\[ \theta' = \frac{\partial \theta}{\partial z} \tag{4.16.37} \]

\[ f_{\text{elastic}} = \frac{1}{2} K_{11} \cos^2 \theta \cdot \theta'^2 + \frac{1}{2} K_{33} \sin^2 \theta \cdot \theta'^2 \tag{4.16.38} \]

Using single elastic constant

\[ f_{\text{elastic}} = \frac{1}{2} K \theta'^2 \tag{4.16.39} \]

The free energy density for the electric contribution is given by [15]

\[ f_{\text{electric}} = -\frac{1}{2} \Delta \varepsilon \varepsilon_0 (\vec{E}, \hat{n})^2 \tag{4.16.40} \]

\[ f_{\text{electric}} = -\frac{1}{2} \Delta \varepsilon \varepsilon_0 E^2 \sin^2 \theta \tag{4.16.41} \]

Combining the elastic and the electric components, the free energy density is given by [15]

\[ f = f_{\text{elastic}} + f_{\text{electric}} = \frac{1}{2} K \theta'^2 - \frac{1}{2} \Delta \varepsilon \varepsilon_0 E^2 \sin^2 \theta \tag{4.16.42} \]

Using the Euler Lagrange’s equation to minimize the free energy

\[ \frac{\delta f}{\delta \theta} = \frac{\partial f}{\partial \theta} - \frac{d}{dz} \left( \frac{\partial f}{\partial \theta'} \right) = 0 \tag{4.16.43} \]
\[-K \theta'' - \Delta \varepsilon \varepsilon_0 E^2 \sin \theta \cos \theta = 0 \tag{4.16.44}\]

When \(\theta\) is small, \(\sin \theta \approx \theta; \cos \theta \approx 1\);

\[-K \theta'' - \Delta \varepsilon \varepsilon_0 E^2 \theta = 0 \tag{4.16.45}\]

Solving, we get the threshold voltage

\[V_c = \pi \sqrt{\frac{K}{\Delta \varepsilon \varepsilon_0}} \tag{4.16.46}\]

Substituting in eq we get

\[-K \theta'' \frac{K \pi^2}{V_c^2} E^2 \sin \theta \cos \theta = 0 \tag{4.16.47}\]

\[\theta'' + \frac{1}{2} \left( \frac{E \pi}{V_c} \right)^2 \sin 2\theta = 0 \tag{4.16.48}\]

Using isotropic condition we get

\[-\nabla^2 \theta + \frac{1}{2} \left( \frac{E \pi}{V_c} \right)^2 \sin 2\theta = 0 \tag{4.15.1.49}\]

Using the Laplace eq in cylindrical co-ordinates.

\[\left\{ \frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial \theta}{\partial r} \right) \right\} + \frac{\partial^2 \theta}{\partial z^2} + \left\{ \frac{\pi E(r,z)}{U_0} \right\}^2 \sin 2\theta = 0 \tag{4.16.50}\]

This equation is solved in the rectangular region

\[0 \leq r \leq \infty \tag{4.16.51}\]

\[0 \leq z \leq d \tag{4.16.52}\]
With boundary conditions that ensures the homeotropic condition on the substrates

\[
\theta (r)|_{z=0} = 0 \quad (4.16.53)
\]

\[
\theta (r)|_{z=d} = 0 \quad (4.16.54)
\]

And the axial symmetry of the system are satisfied

\[
\frac{\partial \theta}{\partial r}|_{r=0} = 0 \quad (4.16.55)
\]

\[
\frac{\partial \theta}{\partial r}|_{r=\infty} = 0 \quad (4.16.56)
\]

After estimating the tilt angle numerically, the phase lag profiles \(\Delta \Phi(r)\) can be calculated from (3.15.1.57)

\[
\Delta \Phi(r) = \frac{2\pi}{\lambda} \left[ \int_0^d n(\theta(r,z))dz - n_0d \right] \quad (4.16.57)
\]

Where the refractive index \(n(\theta(r,z))\) can be determined using eq. (3.15.1.58)

\[
n(\theta) = \frac{n_0n_e}{\sqrt{n_e^2\cos^2\theta + n_0^2\sin^2\theta}} \quad (4.16.58)
\]

\(n_e\) and \(n_0\) are the refractive indices of the extraordinary and ordinary rays

Now to understand what is happening, inside and outside the hole region, it is important to refer to [17] which uses the same PI, SE1211, and the same diluting solvent, NMP as used in my experiments. The LC used also has a negative dielectric anisotropy \(\sim 4.2\) and has initial homeotropic orientation. A high frequency AC voltage is first applied for PI with different thicknesses and allowed to relax. It is found the higher the thickness of the PI more is the time required for the system to relax after the voltage is switched off. It has been suggested that selective
adsorption of ions from the limiting surfaces of the liquid crystal might be responsible for this phenomenon leading to an electric field of ionic origin, localized close to the surfaces on a layer whose thickness is of the order of the length of Debye. The adsorbed ions causes electric-field screening. Other effects like the dependence of the weak anchoring energy on the thickness has also been suggested. It has been mentioned in [17] that the effective anchoring energy has a bulk contribution related to the selective ionic adsorption which is responsible for a surface electric field. The surface energy related to this field is partially due to the dielectric anisotropy of the nematic liquid crystal and partially to the linear coupling of the surface field with the flexoelectric polarization.

Another potential source of ions is the possible presence of impurities in polyimide SE1211. The SE1211 used in my experiments is pre-imidized. However, during the imidization process by the manufacturer, it is quite possible that impurities get accumulated from the unreacted monomers. It has been reported that during the curing of BPDA-PDA polyimides [19], either due to a bad diffusion of temperature or due to the difficulty to remove by-products and water molecules inherent in the imidization reaction, remaining impurities can accumulate and act as ionizable centers supplying free mobile charges. The thicker the coating the more would be the amount of impurities entrapped. Thus we have the possibility of multiple sources of impurities in our case.

The screening effect, as mentioned earlier, is characterized by the Debye screening length [14] which is given by

\[ \kappa^{-1} = \sqrt{\frac{\varepsilon_0 \varepsilon^* k_B T}{2 n_0 q^2}} \]  (4.16.59)

Where \( n_0 \) is the number of ions per unit volume, \( q \) is the charge in C, \( \varepsilon_0 \) is the vacuum permittivity given by \( 8.85 \times 10^{-12} \) C/Vm, \( k_B \) is the Boltzmann constant given by \( 1.3 \times 10^{-23} \) J/K. \( T \) is the
temperature in K. $\varepsilon$ is the relative permittivity of the LC. Using $n_0 = 10^{20}$ m$^{-3}$, $\varepsilon \sim 10$, $T \sim 300 K$, we get $\kappa^{-1}$ as 300 nm. Thus if the Debye screening is 300nm, it means that the potential is effectively screened outside 300nm. In case of field screening the free charges are able to compensate the charges on the electrode, only in case of small fields. When the charges on the electrode are larger than the total free charges, then the material experiences a DC electric field.

The minimum value of the DC voltage can be determined from the threshold electric field as given in [14]. The threshold electric field is given by $E_{th} = \frac{q\cdot n_0 \cdot d}{\varepsilon_0 \cdot \varepsilon}$. Using $q = 1.6 \times 10^{-19}$ C, $n_0 = 10^{-19}$ m$^{-3}$, $d = 50 \mu m$ and $\varepsilon_0 \cdot \varepsilon = 10^{-10}$ C/Vm, we get $E_{th} = 10^5$ V/m. That means for a 50 $\mu m$ cell, we can switch the director using a 5V DC [14]. This value is within the statistical range of values that we have obtained in Table 1. The critical DC voltages that we have obtained for reorienting the LC outside the hole region, are 2.5V, 4.5V, 5.5V and 6 V for increasing thickness and for initial PI:Solvent ratio of 1:8, 1:4, 1:2 and 1:1 respectively. From fig. 3.63 it can be seen that the DC voltage keeps increasing with thickness, while the AC voltage remains constant. The constant value of the AC voltage is due to the average velocity of the ions. The velocity of the ions which migrates towards the electrodes on the application of an electric field is given by eq. 4.16.60.

$$\vec{v} = \mu \vec{E}$$  \hspace{1cm} (4.16.60)

where $\vec{v}$ is the velocity, $\mu$ is the mobility tensor and $\vec{E}$ is the electric field.

If $E_i$ is a component of the electric field then,

$$\vec{v}_i = \mu_{comp} \vec{E}_i$$  \hspace{1cm} (4.16.61)

where $\mu_{comp}$ is a scalar component corresponding to $\vec{v}_i$ and $\vec{E}_i$. 

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The electric field \( \vec{E} \) is a vector having both magnitude and phase and can be given by eq. 4.16.62.

\[
\vec{E}_i = E_0 \sin \omega t \ \hat{k}
\]  

(4.16.62)

The mean of \( \vec{E} \), i.e \( \vec{E}_i \) averaged over time period \( T \), is given by eq. 4.15.1.63.

\[
\langle E_i \rangle = E_0 \int_0^T \sin \omega t \ dt = E_0 \left[ \frac{\cos \omega T}{\omega} - \frac{1}{\omega} \right] = E_0 \left[ \frac{\cos \frac{2\pi t}{T} - 1}{\omega} \right] = 0
\]  

(4.16.63)

Thus for AC voltages, the \( \langle \vec{v}_i \rangle \) is also equal to zero. This implies that the ions will not be affected by the electric field in case of AC, over one time period, while they will be affected in case of DC voltage. The DC voltage thus requires higher voltage to re-orient the molecules and overcome the effect due to field screening. The DC voltage allows movement of the ions and the difference between AC and DC voltage is given in ref [18]. While the average velocity of the ions using AC voltage is zero over one time period, the distance travelled by the ions using DC voltage is given by

\[
d_{DC} = f(\mu)E_{DC}\Delta t
\]  

(4.16.64)

Where \( d_{DC} \) is the distance moved by the ions, \( \Delta t \) is the time for which the voltage is applied and \( E_{DC} \) is the electric field.

Using \( E_{DC} = \frac{V_{DC}}{d} \), where \( V_{DC} \) is the applied voltage and \( d \) is the thickness of the cell,

\[
d_{DC} = f(\mu)\frac{V_{DC}}{d} \Delta t
\]  

(4.16.65)
Now coming to within the hole etched region, with the increasing PI thickness, the number of absorbed ions will also be higher. With the presence of the inhomogeneous electric field, over a 5 minute period, the adsorbed ions under DC voltage will migrate accordingly while they will not in case of AC voltage. Thus inside the hole etched region, a very different trend is observed. The DC “completion voltage” keeps decreasing as the thickness of the PI coating keeps increasing. This can be seen from Table 1. According to the table, for a hole diameter of 1mm, the “completion voltage” is 28V DC for the thinnest PI thickness, represented by initial PI:Solvent ratio of 1:16 and 18V DC for the thickest PI thickness represented by initial PI:Solvent ratio of 1:1. In between for different thicknesses, the DC “completion voltages” are 22V, 23V and 24V represented by initial PI: Solvent ratios of 1:2, 1:4 and 1:8 respectively. Based on what is discussed before, for outside the hole region, a thicker PI will have more adsorbed charged ions. Inside the hole region, more availability of charged particles would mean more migration of them towards the electrodes when DC voltage is applied for a significant period of time of 5 min. This would mean greater polarization and hence for a thicker PI coating, the presence of a larger value of the electric field. This may help to explain why a lower “completion voltage” is required for a thicker PI as compared to a thinner PI for the same hole diameter and the same cell thickness.

The possible migration of the ions and the corresponding polarization resulting in increased electric field for DC, leading to almost planarization of the LC molecules in some region, may explain why a very different behavior of DC phase profiles are observed as compared to AC phase profiles. For 1 mm hole diameter, as seen in fig. 4.20,4.25, 4.30, 4.35 and 4.40 which corresponds to PI thicknesses of 1:1, 1:2, 1:4,1:8 and 1:16, the phase profile in case of DC voltage shows “penetration” at the “completion voltage” while none of that is observed in case of AC “completion voltage”. Similar trend is observed for 2mm hole diameter as seen in fig. 4.44, 3.48, 3.52, 3.56 and
3.60. Also it is important to note, that the phase profiles obtained are not parabolic in nature in our experiments, as parabolic phase profiles are limited to a hole diameter of 600 μm such that the Releigh criterion is satisfied and the deviations from the parabola are confined to only a quarter wavelength. This corresponds to hole diameter: thickness ratio of 12:1. However, the reason we choose 2mm is to get as close as possible to practical applications like the cell phone camera lens [4].
When the hole diameter is increased, the electrodes require a higher voltage to attract the charges and cause polarization (fig. 4.70). As seen from Table 1, for a hole diameter of 1mm, the DC “completion voltage” is 18V. When the hole diameter is increased, the “completion voltage” also increases. Thus for a hole diameter of 2mm, 3mm and 4mm, the “completion voltages” are respectively 38V, 59V and 110V respectively. The migration of charged particles within the PI due to the influence of an external electric field has been explained through the transport model in the subsequent section. The electric field within the hole region of the cell have also been looked into for very small holes in section 3.8.

### 4.17 Transport Model

In order to consider the distribution of charged particles within the PI due to the electric field, we refer to the transport model [20,21].

Two important assumptions:

1) The total numbers of charged particles are conserved. There is no interaction between the ions and there is no generation or recombination of positively and negatively charged particles.

2) The current density \( j \) at the boundaries is zero

\[
j\left( \pm \frac{d}{2}, t \right) = 0 \quad \text{(4.17.1)}
\]
\[ \rho^\pm(x) \sim \exp\left( \pm \frac{\mu^\pm_{xx} \cdot E \cdot x}{D^\pm_{xx}} \right) \]  \hspace{1cm} (4.17.2)

Where \( \mu \) is the mobility of the charge and is a tensor term and is different in different directions.

The electric field \( E \) is constant along the x direction. \( D \) is the diffusion constant and \( \rho \) is the charge density.

Eq. 4.18.2 gives the distribution of the charged particles in the PI along one dimension under a constant electric field.

4.18 Electric field inside a small hole in a capacitor

To investigate the electric field inside a small hole and along the substrate, which is much smaller compared to the size of the hole used in our experiments, a capacitor with an uniform top substrate and a small hole of radius ‘a’ in the bottom substrate is considered [22]. This is shown in fig. 4.69

\[ \Phi = V \]

\[ \Phi = 0 \]

\[ E_0 \]
Figure. 4.69. A capacitor with an uniform top substrate and a small hole in the bottom substrate. Reproduced from [22], p. 35.

To develop the potential for such a system, oblate spheroidal co-ordinate system has been used [22]. The co-ordinate system is a three dimensional orthogonal co-ordinate system that results from rotating a two dimensional elliptic co-ordinate system around the axis of symmetry separating the foci [23]. Thus in the x-y plane it results in a circle of radius a. According to this system, the co-ordinate parameters are \( \eta, \theta, \phi \). These relate to the cartesian co-ordinate system as

\[
x = a \cosh \eta \sin \theta \cos \phi \\
-\gamma = a \cosh \eta \sin \theta \sin \phi \\
z = a \sinh \eta \cos \theta
\]

For a surface of constant \( \eta \) forming oblate spheroids, the trigonometric identity is given by

\[
\frac{x^2 + y^2}{a^2 \cosh^2 \eta} + \frac{z^2}{a^2 \sinh^2 \eta} = \cos^2 \theta + \sin^2 \theta = 1
\]

Similarly for a surface of constant \( \theta \), forming half hyperboloids the trigonometric identity is given by

\[
\frac{x^2 + y^2}{a^2 \cos^2 \theta} - \frac{z^2}{a^2 \sin^2 \theta} = \cosh^2 \eta - \sinh^2 \eta = 1
\]

For positive \( \theta \) the half hyperboloid is above the x-y axis while for negative \( \theta \) it is below the x-y axis. Using this co-ordinate system, the following value of the potential has been reported [22].

\[
\Phi(\eta, \theta) = \begin{cases} 
a. E_0. \sinh \eta \cdot \cos \theta + \frac{a * E_0}{2\pi} [-\sinh(\eta) \cdot \tan^{-1}(\sinh \eta) - 1] \cdot \cos \theta, & 0 < \theta < \frac{\pi}{2} \\
-\frac{E_0}{2\pi} [-\sinh(\eta) \cdot \tan^{-1}(\sinh \eta) - 1] \cdot \cos \theta, & \frac{\pi}{2} < \theta < \pi 
\end{cases}
\]

Where \( E_0 \) is the unperturbed electric field between the plates and \( a \) is the radius of the ho
In eq. 4.8.6, $0 < \theta < \frac{\pi}{2}$ refers to the region above the hole while $\frac{\pi}{2} < \theta < \pi$ refers to the region below it.

The far field potential in the upper region $z > 0$, is

$$\Phi \approx \frac{aE_0}{\pi} + \frac{E_0a^3}{6\pi} \frac{1}{r^2} \cos \theta$$

(4.18.7)

where $r >> a$

4.19 Summary

In summary, the work here presents results based on observation of a phenomenon that has not been previously observed. In particular, presented in this thesis is the observation of a dramatically different behavior of a hole-pattern device under AC and DC fields. Electro-optic characterization results using four hole patterned LC cells of different sizes of the electrode etched region, with the PI having homeotropic alignment, have been studied and the electro-optic response have been compared after application of both AC and DC voltages. The electro-optic response obtained using DC voltage is found to be very different than that obtained using AC voltage. Different PI thicknesses have also been studied and the electro-optic response using DC voltage seems to have a PI thickness dependency which is not found in the AC voltage response. In case of DC, the voltage required for complete re-orientation of the LC molecules inside the hole etched region or “completion” is different from AC. Also the measured phase retardation profile in case of the DC voltage, unlike AC voltage response, has the inhomogeneous region starting away from the edges of the hole thereby showing “penetration” inside the hole etched electrode region for certain voltages. The thicker the PI the lower is the DC voltage for “completion” while for AC voltage it is independent of the PI thickness. Also the hole aperture itself seems to change with the
application of DC voltages in certain cases. For outside the hole etched region, the reverse occurs for DC threshold voltage with respect to the PI thickness dependency while it is independent of the PI thickness for AC voltages. Overall, the DC voltages, be it the threshold voltage outside the hole region or the “completion” voltages inside the hole region is always higher than AC voltage for the same hole diameter and PI thickness. This phenomenon in which the electro-optic response using DC voltage is dependent on the PI thicknesses was never reported before and has been reported for the first time in this dissertation. The differences in the electro-optic responses between AC and DC voltages has also been reported for the first time in this dissertation.

Several possible factors like adsorption of ions, PI thickness, and screening may contribute to the differences in electro-optic behavior between AC and DC voltages. The fact that under AC voltage the ions do not migrate over a time period average while they do over time under DC voltage, enormously affects this difference. As a simplistic model the migration of charged particles in the PI in one dimension under a constant electric field have been illustrated. The nature of the electric field, which causes the migration of charges to happen, inside a small hole in a capacitor has also been explored.

As a future extension of the research work done here, cells can be fabricated with a ratio of up to 3.5, between the hole diameter to the thickness of the cell in order to obtain a parabolic phase profile as seen in lenses as given in [Ch 3, 4]. This may help to create lenses with DC voltage or hybrid lenses using both AC as well as DC. Lens characteristics like focal length, aberrations, lens resolution and surface quality can also be measured to characterize the lens. These lenses may have special properties like adjustable aperture while using DC voltage exclusively or as a hybrid of AC and DC voltage. Due to constrain of time and resources, we have not been able to carry out this work which can be carried out as an extension of this dissertation work.
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CHAPTER 1

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CHAPTER 2


CHAPTER 3


CHAPTER 4


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Appendix
CHAPTER 1

Random Lasers

For the experiments described in this chapter the samples have been prepared by me. The filling of cells have been done by me and I have assisted Dr. Antonio Munoz with the experiments. In this chapter, taken from ref [1], an article previously published by me as a co-author, using same patterned cells and the same liquid crystal material, the reverse mode "laser OFF → laser ON" electro-optical switching, upon corresponding electric field states OFF → ON has been investigated. This has been done for different mixtures of nematic liquid crystal Host G (Δε < 0) and PM 597 laser dye. In homeotropic liquid crystal cells, the effects of (i) the dye concentration and (ii) the applied AC voltage on the characteristics of the emission spectra of our samples were analyzed, under ns pulsed laser excitation at 532nm. Although our samples were in the laser OFF state in the absence of an electric field across the cells, and only amplified spontaneous emission (ASE) was observed as a function of the input energy; electrically controlled gain/loss ratio (upon the application of electric voltage of 4-18 V) turned the systems into a laser ON state were the intense, directional random laser (RL) emission occurs, and was characterized. All these results were compared with the observation of RL emission in hometropic, large aperture, hole pattern liquid crystals cells (75μm thick, 2 mm hole diameter) in order to get a better understanding on the effects of the light phase modulation, induced by an inhomogeneous electric field, on the RL characteristics (laser modes and threshold) of our samples.
1.1 Introduction

The last 15 years were marked by a growing interest in lasing using dye-doped liquid crystals with the aim to build a compact, all-organic, tunable mirror-less laser. The potential applications of such lasers range from miniature spectroscopic and medical tools to large area holographic laser displays. Two types of major lasing mechanisms have been developed for coherent light amplification using liquid crystals. They are (i) Band edge cholesteric liquid crystal lasers [1-3] and (ii) random lasers [4-5]. The principle of operations of these two mechanisms and the latest technological advancement in these areas are available in the literature [1-17]. In both cases, to make technological applications possible, important questions have to be addressed. For Cholesteric Liquid Crystal (CLC) band edge lasers, apart from the optimization of Continuous Wave (CW) [12], important parameters like the pulse emission performance, the efficiency of the photon to photon conversion and the design modelling of high-power-tunable output lasers are critical and needs to be addressed. On the other hand for random lasers based on DDLC, it is important to achieve controllable emission. The effect of electrically controlled RL in dye doped nematic liquid crystals having positive anisotropy has been reported [18]. However RL emission characteristics with dye doped liquid crystal having negative dielectric anisotropy has not been reported yet.

This paper deals with the spectroscopic characteristics obtained by applying homogenous and inhomogeneous electric field to a LC cell, filled with LC, Host G, having negative dielectric anisotropy (Δε<0) and having different concentrations of PM-597 laser dye. In the OFF state
(V=0), Amplified Spontaneous Emission (ASE) was detected in the transmissive state of the samples. While in the ON state at V> \( V_{TH} \), i.e with Voltage above the Freedrickz transition threshold, random lasing (RL) occurs for homogenous and inhomogeneous electric fields. The results of experiments suggest that the lasing characteristics of the random laser (threshold, intensity and wavelength) can be optimized by optimizing the applied voltage and the dye concentration of the liquid crystal mixture.

1.2 Sample Preparation and Experimental Setup

The samples used are filled with nematic LC, commercial name Host G (Jiangsu Hecheng Chemical Material Company). The LC is stable between -40 degrees and 110 degrees. The LC parameters are birefringence \( (\Delta n) = 0.150 \) and negative dielectric anisotropy \( (\Delta \varepsilon) = -5 \). In the samples used the LC is doped with Pyrromethene 597 dye. This dye is also referred to as PM597 commercially and is obtained from Exciton. In the samples used is doped with 0.2, 0.5, 1 and 2 wt% of PM597 respectively. The order parameter "S" of PM597 in Host G was verified by polarized absorption experiments in twin cells consisting of parallel glass plates coated with rubbed polyimide alignment layers, to induce a planar alignment. Glass spacers mixed with an UV-epoxy provided cell thicknesses \( \approx 12 \) \( \mu \)m. The cells were capillary filled with pure Host G NLC, in one side of the cell, and with specific dye concentration LC mixture (Host G +0.2 wt% PM597) in the other side (figure 1b). Observing the samples under a polarized microscope showed an average planar alignment, with the optical axis situated in the plane of the cell parallel to the rubbing direction.
The random laser experiments were carried out with conventional sandwich cells consisting of two parallel glass substrates plates separated by Mylar spacers, having thickness of 75 μm. Onto the glass substrates, which are pre coated with a transparent conductive film made of ITO (Indium Tin Oxide), was deposited a thin homeotropic alignment film (PI) of polyimide SE-1211 (Nissan Chemicals Industries Ltd, Tokyo, Japan) by using the following procedure: The commercial PI mixture, polyamic acid SE-1211 (6% by weight) in a solvent (54% N-Methyl-2-Pyrrolidone and 40% Butylcellosolve), was diluted further using "Solvent AMI-26", which consists of 60% N-Methyl-2-Pyrrolidone and 40% Butylcellosolve. This solution was spin coated onto the ITO film at 500 rpm for 10 s and then at 2000 rpm for 20 s. The substrates were prebaked at 100 C for 3 min to evaporate the solvent and then baked at 190 C for 30 min for complete imidization, as prescribed by the manufacturer, to induce a homeotropic alignment of NLC molecules at the interface. The cell was capillary filled with the flow direction along the rubbing direction. Upon observing the samples under a polarized microscope, they showed homeotropic alignment, with the optical axis situated perpendicular to the plane of the cell. For the optical and spectroscopic characterization of our LC samples, different configurations of our basic experimental setup (figure 1a) were employed.
Figure 1: (a) Experimental setup. Configurations A and B are defined by the light source, the sample position and detector employed in each case, as described through the text. The linear polarizer, the beam splitter (BS), and the focusing lens are common to all configurations. (b) Polarized absorption band of PM597 in Host G NLC. The twin planar liquid crystals cells are rubbed in the direction indicated with the arrow (inset). The relative orientation of the sample with respect to the polarization of the incident light has been identified as follows: P= Rubbing direction parallel to the incident electric field and PP= rubbing direction perpendicular to the incident electric field. Taken as it is from reference [1]
As light source we employed (i) an Ocean Optics LS-1 Tungsten Halogen lamp (for the order parameter determination), (ii) an air cooled Ar CW laser, Ion Laser Technology model 5405A (for the light transmission vs voltage experiments at 477nm) and (iii) either a tunable pulsed OPO Nd:YAG Laser, Opolette 355II +UV (OPOTEK Inc.) with 5ns pulse duration or a frequency doubled Coherent Nd:YAG laser with 8ns pulses at $\lambda = 532$nm (for the random laser experiments). As detectors were employed: (i) Either an Ocean Optics HR2000 spectrometer (with $<1$ nm spectral resolution) or a Jobin Yvon-Spex TRIAX 550 spectrometer fitted with an i-Spectrum ONE intensified CCD detector operated in the continuous mode, for the random laser experiments (Spectrometer 1) and for the order parameter determination (Spectrometer 2), (ii) a Newport Research Power Meter model 815, for the light transmission vs voltage experiments at 477nm (Head 2), and (iii) a Molectron Optimum 4001 energy meter or a RJ-7620 Energy Radiometer, Laser Precision, (Heads 1 and 2) to measure the pulse input laser energy at 532nm, in the random laser experiments. In our experimental setup (figure 1a) the polarizer, the beam splitter (BS) and the focusing lens (20cm focal length) are common in all the configurations while the samples were set in position B only for the random laser experiments
1.3 Results and Discussion

1.3.1 Planar Liquid Crystal Cells

Due to its importance for the design and engineering of new systems for controllable laser emission based on liquid crystal materials doped with highly fluorescent dyes, special attention has been given to the evaluation of the orientation of the dye molecules dissolved in the nematic liquid crystalline phase [19]. By means of polarized absorption, the order parameters of the dyes in the liquid crystal mixture can be determined. With our samples in position A (figure 1a) we measured the absorbance spectra of PM597, relative to the pure Host G NLC, for two different geometries: (a) When the polarization of the input white light is parallel to the rubbing direction of the thin planar liquid crystal cell (A∥) and (b) when the polarization of the input white light is perpendicular to the rubbing direction of the planar cell (A⊥). In our setup the polarization of the light is kept vertical and by rotating the sample we could make both geometries. When the dye molecules are parallel to the polarization of the incoming light we observed the sample to have the maximum absorption. (continuous line in figure 1b). The order parameter, S, of the dye molecules was estimated to be $S = (A_\parallel - A_\perp)/(A_\parallel + 2A_\perp) = 0.48 \pm 0.01$. This value is higher than previous results reported in the literature [20, 21], and corresponds to the PM597 dye in the host G NLC, at room temperature.
1.3.2 Homeotropic Liquid Crystal Cells. Random Laser Experiments

1.3.2.1 The OFF State at V=0 Volts

For the random laser experiments, the homeotropic samples were set in position B (Figure 1a); with the rubbing direction parallel to the polarization of the pump beam. The light excitation was incident at \( \sim 10^\circ \) from normal to the plane of the cell. A multichannel CCD spectrometer with a fiber termination was used to capture the emission spectra (Spectrometer 1); and the applied voltage was controlled with a power supply. Figure 2a shows, for the sake of comparison, the characteristic emission band observed from sample AMI-1(Host G + 0.5wt% PM597) under CW (Dotted pink line) and pulsed laser excitation (Continuous red line) at 532nm in the OFF state (Transmissive mode), at V= 0 Volts.
Figure 2: (a) Characteristic emission band observed from sample AMI-1(Host G + 0.5wt% PM597) under CW (dotted pink line) and with pulsed (≈ 0.55 mJ/pulse) laser excitation (continuous red line) at 532nm in the OFF state, at V= 0 Volts. (b) FWHM and Emission intensity (inset) vs. pump energy for the Amplified Spontaneous Emission (ASE). Taken as it is from reference [1].

At low input powers our samples are characterized by a broad emission band with Full Width at Half Maximum, FWHM, ≈ 50nm, and maximum at $\lambda_{\text{MAX-Fluor}} \approx 577$nm. These observations are in good agreement with previous characterizations of the fluorescence associated with PM597 in
NLC [20], which has been attributed to the spontaneous transition from the first excited state $S_1$ to the ground state $S_0$. When the pump energy was increased, the emission band narrowed as a function of the input power to a FWHM $\approx 5$nm (figure 2b). Due to ground state absorption (GSA), there is a red shift between the maximum, $\lambda_{\text{MAX-ASE}}$, of the narrow stimulated emission observed at $\lambda_{\text{MAX-ASE}} \approx 587$nm, as compared with $\lambda_{\text{MAX-Fluor}}$ of the peak fluorescence emission of the samples at low input powers (figure 2a). Previous observations in systems with PM597 laser dye dissolved in nematic liquid crystals with negative dielectric anisotropy indicate that, in cells with homogeneous alignment, random laser modes are the main characteristic of the emission spectra, for pump energies above certain threshold [22]. At variance with those results, the data portrayed in figure 2b (inset) show an almost linear enhancement of the output intensity, owing to preferential amplification at frequencies close to the maximum of the gain spectrum. No clear threshold, coherent directional emission, nor laser modes are present in the emission of our samples at $V=0$ Volts, within the limits of our experimental setup. These observations constitute an important difference with respect the data reported in the literature [22] and accounts for the presence of poorly balanced gain/loss ratio in the transmissive OFF state of our AMI-1 systems.

1.3.2.2 The ON State at $V > 0$ Volts

A) Homogeneous Electric Field.

Controlling the liquid-crystal (LC) alignment by external fields is the key principle in many LC photonic devices. As an electric field is applied to the LC cell, it results in a torque to the dipole moment of the LC molecules. In our systems, with hometropic alignment, the molecules of the liquid crystal (with negative dielectric anisotropy) are aligned perpendicular to the applied electric
field, introducing an elastic deformation of the system, in order to minimize its Gibbs free energy. This deformation is characterized, as a function of the applied voltage, by the "tilt angle, \( \theta(z) \), of the director, measured relative to the plane of the glass substrates. Starting with a value of \( \theta(z) \approx 90^\circ \) in the transmissive mode, at \( V=0 \) volts, for a sample positioned between crossed polarizers, the polarized light produced by the first polarizer travels through the sample unchanged and is subsequently absorbed by the second polarizer. Therefore, extinction occurs, and the cell appears dark (Figure 3a).

![Figure 3: Light transmission of sample AMI-1 between crossed polarizers at (a) V= 0; (b) after 1 min at V= 16 Volts and (c) after 20 min at V= 16 Volts. (d) Absorbance (at \( \lambda_{\text{ex}} = 477\text{nm} \)) vs Voltage. (e) Emission spectra and light intensity at 578nm (inset) vs voltage. The results presented in (d)
and (e) correspond to experiments that were performed in our samples after 20 minutes at the applied voltage indicated in the figure. Taken as it is from reference [1].

At $V > V_{TH}=1.8\,\text{V}$ ($V_{TH} = \text{threshold voltage}$) the tilt angle of NLC molecules is larger near the substrates and has a minimum at the center of the cell, where the host LC will have the tendency to be oriented parallel to the plane of the glass substrates (absorptive mode). This molecular reorientation induces a phase retardation $\Delta\gamma$, and allows some of the incident light to traverse the second polarizer. In switching the cell to the ON state, topological defects will also appear, as consequence of going from homeotropic $\rightarrow$ homogeneous symmetry. The relaxation time $\tau_{rel}$ depends on the applied electric field, the specific characteristics of the host, and on the geometry of the cell. In our cells, the relaxation time of the defects induced by the electric field was observed to be $\tau_{rel} \geq 20\,\text{minutes}$, when the voltage was in the $4 < V < 16\,\text{Volts}$ range (figures 3b and 3c). Therefore all our observations on the effects of the electric field on the optical properties of our samples were always investigated at $t \geq 20\,\text{min}$. At $V > V_{TH}\,\text{Volts}$, the reoriented molecules (parallel to the substrate of the cell) of the LC mixture will be responsible for enhancing the light absorbance, within the absorption band of the laser dye (figure 3d). In our samples, the basic mechanism for RL is coherent backscattering [5, 23, 24], expected to be strongly influenced by gain. Then, the effects of the electric field on the optical response and the spectroscopic characteristics of the emission spectra of our sample AMI-1 were analyzed at different voltages. The main results are portrayed in figure 3e. Figure 3e shows, as a function of the AC voltage, the emission spectra of sample AMI-1, under constant input laser excitation of $\approx 500\,\mu\text{J} / \text{pulse}$, at 532nm. At low voltage ($V \leq 4V_{TH}$) the emission spectra show the same narrow emission band of the PM597 dye, previously observed in the OFF state of our samples (Figure 2a). Upon increasing the applied voltage (at constant input power laser excitation), a slight blue shift in the peak
emission was observed, similar to the one reported for the Amplified Spontaneous Emission (ASE) in nematic liquid crystals doped with pyrromethene dyes [25]. On the other hand, at $V \geq 6V_{TH}$, discrete sharp peaks emerge at the blue tail of the emission band, and a bright spot (typical of random lasers) appears on the background screen. The intensity of these sharp peaks changed as a function of the applied voltage (figure 3e), while the wavelength of the maximum peak emission ($\lambda_{RL} \approx 575\text{nm}$, at $V= 12\text{Volts}$) and the line width (FWHM $< 1\text{nm}$) of these sharp modes are in good agreement with, previous reports of the random laser emission characteristics in homogeneous liquid crystal cells, filled with mixtures of nematic liquid crystals with negative dielectric anisotropy, and PM597 laser dye [22]. All these results, combined with data reported in the literature [22], suggest that the discrete spectral modes shown in Figure 3e result from spatial resonances in the liquid crystal mixture, and that such resonances are related to the configurations of the liquid crystal mixture AMI-1 in the "ON" state, at different AC voltage. The inset in figure 3e portrays the variation of the peak intensity at 578nm (random laser), as a function of the input laser energy at 532nm, at constant applied voltage of 16 volts. In Figure 3e (inset), a nonlinear increase with increasing the pumped energy, and a clear energy threshold ($E_{th} \sim 420 \mu\text{J/pulse}$) can be obtained, which is a confirmation for the occurrence of a random lasing emission. The lasing threshold value estimated from figure 3e for the onset of the random laser emission in our sample AMI-1 is higher than the one reported in the literature, for homogeneous liquid crystal cells, filled with a similar mixture of a nematic liquid crystal with $\Delta\varepsilon = -5$, 0.5 wt % PM597 [22]. In addition, the fact that, under our experimental conditions (beam waist at the sample, and input pulsed laser energy $\leq 0.6 \text{mJ}$ at 532nm) above the lasing threshold, the emission spectra of the sample AMI-1 present, simultaneously, the random laser emissions and the ASE band peaking at $\approx 587\text{nm}$, indicates that further optimization of the system has to be done in order to (i) improve the balance
gain/loss, (ii) to maximize the lasing efficiency and (iii) to low the lasing threshold. In order to analyze the effect of the dye concentration on the random laser characteristics of our LC mixtures, samples with lower and higher wt% of PM597 in Host G were prepared (mixtures AMI-2 and AMI-3, respectively). In Figure 4a the emission spectra of sample AMI-2 are portrayed at \( V = 0 \) Volts (pink dotted line) and \( V = 4 \) Volts (Red continuous line). The spectra in figure 4 were recorded under a constant laser excitation of 0.2 mJ / pulse, at 532nm. At \( V = 0 \) Volts (OFF state), the emission spectra of AMI-2 follows a similar trend (Line narrowing and linear increase of the ASE as a function of the input energy) that the one observed before for sample AMI-1 (figure 2). However, at \( V \geq 4 \) Volts (ON state) a field induced ASE → RL conversion occurs (at constant input laser energy), as is shown in Figure 4a (red continuous line).
Figure 4: (a) Emission spectra of the sample AMI-2 under pulsed laser of 0.2mJ/pulse at 0 (pink dotted line) and 4 (red line) Volts. (b) Random Laser threshold vs Voltage in sample AMI-2, and lasing threshold vs the dye concentration in samples AMI-1, AMI-2 and AMI-3 (inset) at V= 16 Volts. Taken as it is from reference [1].
Moreover, no evidence of the ASE emission band was clearly identified in the ON state, within the resolution of our experimental setup, for input energies above the lasing threshold. In the ON state, the field induced absorption of the dye improves the gain/loss ratio in sample AMI-2, and a reduction of the lasing threshold was observed, as a function of the applied electric field (Figure 4b). At higher dye concentrations, evidence of electrically controlled switch ON random laser in sample AMI-3 was also observed. For the sample AMI-3 in the OFF state ASE emission was detected, while in the ON state the optical response of the sample showed characteristics similar to the sample AMI-1, as a function of the input power (Figure 3e). Looking at the effect of the applied electric field on the lasing characteristics of the samples AMI-1, AMI-2 and AMI-3 we observed that, in general (i) no RL emission occurs in OFF state of our samples; (ii) at $V > V_{\text{THR}}$ the lasing threshold decreases as a function of the applied electric field and (ii) at fixed voltage, $V=16$ Volts, RL occurs at the lowest threshold in sample AMI-2 and increases with the dye concentration in our samples (Figure 4, inset). To the best of our knowledge, all these results represent the first report of electrically controlled OFF (homeotropic) $\rightarrow$ ON (planar) random laser, based on a liquid crystal host material with negative dielectric anisotropy.

**B) Inhomogeneous Electric Field**

Above, switchable random laser emission was obtained in homeotropic liquid crystal cells filled with dye doped host G. The spectral characteristics (FWHM, peak emission wavelength and lasing threshold) of the emission observed in our samples show a strong dependency on the applied electric field, due to the delicate balance between gain and the feedback mechanism (light scattering by the host), modulated by the electric field. In fact, while at $V=0$ Volts no clear
indication of lasing has been observed in any of our samples with hometropic alignment (within the range of input laser energy analyzed); whenever the condition \( V > V_{TH} \) was satisfied, clear evidence of random laser emission has been presented, for input energies above the corresponding lasing threshold. All these results combined with those reported in the literature indicate that the random laser emission depends on the degree of reorientation of the liquid crystal mixture molecules under the external electric field. In RLs, an important parameter which affects the lasing threshold is the transport mean free path, \( l \), which is defined as the distance a photon travels before its direction is randomized [26, 29]. The light-scattering cross sections, which originate from local director fluctuations [30], depend on the directions of the electric field and the propagation vectors with respect to the average molecular orientation. As a result, the magnitude of \( l \) can be controlled directly by the amplitude of the applied electric field as the sample passes through different scattering states. On the other hand, the reorientation effect of nematic molecules in an inhomogeneous electric field, created by a special electrode geometry, is used also to control the phase, \( \Delta \phi \), of the light wave [31, 33]. To produce this modulation in \( \Delta \phi \) with a LC layer of uniform thickness, it is essential to design an electrode system capable of creating an axially symmetrical and spatially inhomogeneous electric field. The desired electric field distribution can be formed, in its simplest approach, by using electrodes in the glass LC cells, with circular holes. The results of experimental studies of the electro-optic properties of nonsymmetric hole pattern liquid crystal cells with homeotropic orientation has been reported [34], and motivated to explore the effect of such inhomogeneous electric field, on the random laser characteristics of dye doped nematic liquid crystals. In this case, our hole pattern LC cells were constructed such that there is an electrode with a hole in the center, of diameter \( L=2 \) mm, made by etching the conductive ITO film on one
of the glass substrates. The second electrode is a transparent layer of indium oxide, deposited on the second glass substrate of the cell.

Figure 5: Crossed polarizers transmission observed in sample AMI-4 at (a) V=0 and (b) V=16 Volts, respectively. (c) Phase-lag profile at V= 16 Volts. (d) Emission spectra in the OFF state (black) and in the ON state at V=16 volts (R1, pink dotted line; R2 red continuous line). Taken as it is from reference [1].

In order to contrast the electro optical properties of our hole pattern liquid crystal cells with those presented in previous sections, in the case of the cells with asymmetric electrodes the surfaces of the substrates in contact with the LC layer were covered with an homeotropic alignment layer (polyimide 1211); rubbed and filled with a mixture of Host G NLC doped with PM597 laser dye.
(2%wt, sample AMI-4). Two regions, R1 and R2, may be identified in our hole pattern LC cells. The region R1 corresponds to the volume of the cell where the applied electric field is perpendicular to the plane of the glass substrates (homogeneous electric field region); and the hole region R2, where the field is stronger at the edge of the opening, and decreases gradually from the edge to the center of the hole area. Since the LC possesses negative dielectric anisotropy, the nematic molecules tend to turn across the field. As a result, a radially symmetric refractive index distribution is formed as the voltage is gradually increased. In Figure 5b the interference pattern observed in the sample AMI-4 (at V= 16Volts) through a polarization microscope is portrayed. Inside of R2 the reorientation of the LC molecules is appreciable and a number of interference rings are observed in the microscope with crossed polarizers. In figures 5b and 5c the center of R2 looks dark, and the phase profile (Δφ) has a plateau, respectively, due to the homeotropic alignment of the molecules in the LC mixture. In good agreement with the results presented above, the emission spectra associated with the OFF state of the hole pattern LC sample show the characteristic ASE portrayed in Figure 5d (black dotted line). However, in the ON state of the sample at V = 16 Volts, RL emission was observed with different spectral characteristics in the output signals (figure 5d) of AMI-4 after pumping the sample at R1 (pink dotted line) and at R2 (continuous red line). From this figure, in the OFF state, the LC alignment is homeotropic and the dye molecules tend to follow the LC. The absorption of the dye molecules and the gain in the system has a minimum in this configuration and hence only ASE (5nm FWHM) was detected, peaking at 587nm. When an AC voltage of 16V is applied to the cell, the LC molecules in R1 are expected to be planar. This is because the voltage applied is above the threshold transition voltage and the LC has negative dielectric anisotropy. Since the dye molecules are expected to follow the LC molecules, the
absorption of the polarized light will be along the dipole moment of the dye molecules and is expected to be maximum. The emission results in various random lasing modes (Pink dotted line in figure 5d), with the maximum lasing intensity at 572nm. In Figure 5d (insets), the green light corresponds to the transmitted input pump beam at 532nm and the yellow light is associated with the output random laser signal. The output random laser signal is highly directional and the detector needs to be placed perpendicular to the LC cell in order to obtain the signal. The random laser emission coming out from the hole region R2 presents interesting results, compared with the observations at the region R1. They are summarized in figure 5d (continuous red line).

The comparison of the spectral characteristics of the lasing signal associated with the regions R1 and R2 in our hole pattern liquid crystal cell reveals a red shift in the wavelength of the output random laser signal as a result of the spectral dependency of the scattering efficiency associated with the different liquid crystal orientations in these two regions of the cell. Diffusive lasing occurs in dye doped nematics by recurrent light scattering and the lasing frequencies are determined by phase relationship between counter-propagating scattered light waves. In general, the blue shift of the laser emission with respect to the fluorescence maximum is determined by interference effects which introduce coherence and feedback, leading to lasing action [5]. The red shift observed in the laser emission in region R2, compared with the one coming out from region R1, reveals differences in the interference effects occurring in these two regions, probably as a result of the phase modulation of the light, produced by the liquid crystal in the region R2. Currently experiments are being conducted in our laboratory in order to corroborate this speculation and to optimize the performance optical of our, electrically modulated, random laser and light phase modulator combination.
1.4 Conclusions

We observed and characterized electrically controlled random laser action in uniform and hole pattern liquid crystal cells filled with a new dye-nematic liquid crystal composite. Experimental results show that the externally applied voltage on the cell and dye concentration can control the lasing intensities of the generated random lasers and their energy thresholds. Light phase modulation induced by inhomogeneous electric fields resulted in a red shift, of the random laser emission observed in our hole pattern liquid crystal cells. To the best of our knowledge all these results are the first report of reverse mode switching OFF→ ON of the random laser emission in dye doped liquid crystals under homogeneous and inhomogeneous electric fields.

Acknowledgments.

This research was supported in part by CONACyT, Mexico. The authors will like to thank Prof P Palffy-Muhoray for making the facilities of his laboratory at the LCI available and to ArteHiga for the assistance in the art of the figures.
Bibliography


CHAPTER 2

Micro-fluidics

For the experiments described in this chapter the samples have been prepared by me and the experiments have been done by me. In the dissertation, after confining the liquid crystal within a particular geometric shape and by using a patterned electrode, the electro-optic response of the system has been studied. Instead of using a confined geometric shape, the LC material can be captured standalone as Liquid crystalline elastomer (LCE) droplets. Future possibilities include study of the electro-optic properties of these droplets. The droplets are generated using micro-fluidic systems. In this chapter, taken from ref [1], an article previously published by me as a co-author, such a micro-fluidic system has been described and the study of the optical properties of the droplets under stress have been elaborated.

1.1 Introduction

LCEs are unique materials combining rubber elasticity and liquid crystalline orientational order [2–3]. The coupling of orientational order and polymer network elasticity results in anisotropic elastic properties and allows for controlled deformation of liquid crystalline order by applying mechanical stress. Following the pioneering work of Finkelmann et al. [4, 5] many nematic LCEs have been developed over the years [6–9]. Until recently, LCE samples described in the literature were of macroscopic size (hundreds of micrometers to several centimeters), mainly in the form of films or, in a limited number of cases, fibers [10–17].
Fig. 1. Compounds used for LCE precursor: reactive mesogenic monomer (1), benzophenone photo-cross-linker (2), cholesteric monomer (3), and poly-hydroxy-methylsiloxane (PHMS). Taken as it is from reference [1].

Fig. 2. A schematic of the microfluidic setup used to produce the LCE droplets. A: Outer phase composed of 70 vol% water and 30 vol% glycerol with 0.36M Tween 80. B: inner oil phase
containing 0.1 wt% of the LCE precursors in chloroform. C: LCE microparticles cross-linked by UV. Taken as it is from reference [1].

In recent years, various methods to produce smaller LCE particles, shells, and rods on micrometer scales have been developed, with a view towards applications as micro-actuators for micro-robots, micro-pumps, and sensors [8, 18, 19]. These micro-actuators respond to external stimuli by changes in shape and/or size. Using a soft lithographic technique, R-responsive anisotropic shape-morphing LCE microrods containing plasmonic gold nanorods [20] and a light-responsive microarray of azo-benzene liquid crystal polymer (LCP) [21] were generated. Zentel’s group adopted microfluidic techniques to generate thermo-responsive LCE microparticles and microshells [22–24]. The average size of these particles and shells ranges from 280 μm to 600 μm, which are ideal for some applications such as micropumps [24] but may be too large for other applications.

To generate smaller LCE particles using microfluidics, one needs to increase the velocity of the outer fluid during the droplet production [25]. However, it is almost impossible to increase the velocity of highly viscous fluids because that would require micropumps to generate pressure higher than their capability [26].

In this paper, we present a method to produce monodisperse LCE microparticles smaller than 100 μm, via oil-in-water (O/W) emulsion templates formed by glass-capillary microfluidics [27–29]. The glass capillary method has been used for the formation of biogels [29], polymersomes [30–32], and porous microparticles of synthetic polymers [33]. However, to the best of our knowledge, the method has not been utilized for the formation of LCE microparticles until now. We demonstrate the production of LCE microparticles and examine the development of nonuniform alignment due to the deformation of these microparticles. Using a finite element
analysis, we find that the director, the average direction of liquid crystal alignment, follows the 1st principal strain field of the sample.

1.2 Experimental

The side chain LC polymers used to prepare the droplets were synthesized according to the procedures described in Refs [2, 34]. LC polymers are synthesized by reacting the terminal vinyl groups of the mesogenic monomer (1, 0.7 mmol), photo-crosslinker (2, 0.04 mmol), and cholesteric monomer (3, 0.26 equivalents), with the Si-H group of poly-hydroxymethylsiloxane (PHMS, 1.0 mmol), via a platinum catalyzed hydrosilylation reaction (Fig. 1). The benzophenone group of the photo-cross-linker [2] will be utilized to induce cross-linking reaction after forming droplets of LC polymer solution (see the following). The pitch (P) of the LC polymer is estimated to be ~ 200 nm, as $P \approx 1/(Ch \times HTP)$, where Ch, the fraction of the cholesteric monomer [3], is 0.26 and HTP, its helical twisting power, is 0.02/nm [35]. PHMS, purchased from ABCR Chemicals, has an average degree of polymerization of ~70 units.

The components are dissolved in thiophene free, dry toluene (200 mL), in which approximately ~30μL of platinum catalysts solution (1% w/w Pt(II)Cl₂ in CH₂Cl₂, Wacker Chemie) is added, and the mixture is stirred at 60℃ in darkness for >18 hrs. Reaction progress is monitored using infrared (IR) spectroscopy by following the 2164 cm⁻¹ peak corresponding to νSi–H, the stretching mode frequency of the Si-H bonds, which decreases in intensity as the reaction progresses. If the reaction is not complete after approximately 18 hours, additional catalyst (~30 μL) is added and the reaction is allowed to continue until the 2164 cm⁻¹ peak decreases to less than
5% of its original intensity. After the reaction is complete, the LC polymer is purified by precipitating the polymer in ice-cold methanol. Finally, the solvent is removed by dispersing the polymer material in benzene, freezing, and sublimating under vacuum, resulting in a whitish, powdery residue. The resulting LC polymer is used as is to prepare LCE droplets.

In order to generate micron sized LCE particles, we first generate emulsion droplets of a LC polymer solution in an aqueous surfactant solution using a glass capillary microfluidic setup, shown in Fig. 2, which consists of a tapered circular microcapillary and a square capillary as described in Refs. [27–29]. The LC polymer solution is pumped through the inner circular microcapillary tip, while the outer aqueous fluid is pumped through one end of the square capillary. We note that the glass capillaries are inert to the volatile solvents which could not be used in previous PDMS and PMMA microfluidic setups, such as those described by Huang et al. [36, 37] and Zhang [38]. The use of organic solvents necessitated the use of glass capillaries, as organic solvents may swell and break microfluidic devices that are made of polymers.

Square glass capillaries (inner dimension: 1.0 mm x 1.0 mm, and outer dimension: 1.4 mm x 1.4 mm) were purchased from VitroCom, Inc., and circular capillaries (1mm outer diameter, 0.58 mm inner diameter) from World Precision Instruments. The inner diameter of the microcapillary tip is ∼20 μm. We fabricated the microcapillaries by first pulling the 1 mm circular capillary with an automated microcapillary puller (P-97, Sutter Instrument), and then breaking the tapered capillary at a point with the desired diameter using a scoring tile (CTS Ceramic tile, Sutter Instrument). The sharp end is then heat trimmed and smoothed with a microforge (MF-830, Narishige International). The glass capillaries are connected though a 27G syringe needle and a polyethylene microtubing (BB31695-PE/2, Scientific Commodities) to gas-tight glass syringes
(1005 TLL, Hamilton Co.) and the flow of each phase is controlled by separate microsyringe pumps (NE-1010, New Era Pump Systems Inc.).

We set capillary flow parameters as follows: the flow rates of the inner and outer phase are 1 ml/hr and 9 ml/hr, respectively. The inner oil phase contains 0.1 wt% of the LC polymer in chloroform (Sigma-Aldrich). The outer phase is composed of 70 vol% water and 30 vol% glycerol (Sigma-Aldrich) with 0.3M Tween 80 (Sigma-Aldrich). House-deionized water is further purified by a Barnstead Nanopure water system, resulting in resistivity >17 MΩ·cm. The purified water is used in all experiments.

The device is placed on an upright microscope (BX-51, Olympus Microscope) with a 10x objective lens to observe the production of emulsion droplets. The initial diameter of the droplets is ~200 μm. They are collected in a long square capillary and are cross-linked under a 15-Watt tabletop UV lamp (peak wavelength = 365 nm) for 1 h. UV radiation activates the benzophenone group of the cross-linker [2] on the LC polymer, and the activated groups react with other LC polymer chains, forming networks. After cross-linking, we check the LCE particles in the long square capillary from different sides, to confirm that they are spherical and do not stick on the surface of the square capillary.

Then, the LCE microparticles are placed between two glass slides and are observed on a cross-polarized microscope. The distance between the two glass slides is varied to compress the LCE microparticles in the z-direction, inducing axisymmetric radial stretching. The birefringence of the deformed microparticle is observed with a full wave retardation plate (λ = 530 nm) in order to resolve the orientation of mesogens in the microparticle.
1.3 Results and Discussion

All the results reported here are obtained after the UV crosslinking. Fig. 3 shows monodisperse microdisks that are confined between two glass slides. The microdisks display a characteristic X-shaped birefringence patterns between crossed polarizers when viewed with a polarizing microscope. To probe how compression of the microparticles affects the pattern, we varied the distance between two glass plates. Without any compression, a typical microparticle has a polydomain structure, consisting of randomly oriented nematic domains (Fig. 4a). If the particle is completely dried, the diameter should be $\sim 20 \mu m$ because the initial diameter is $\sim 200 \mu m$ and the initial concentration of the LCE precursor is $\sim 0.1\%$. However, the average diameter of the particles before any deformation (Fig. 4a) is around $70 \mu m$, implying that they are still significantly swollen by the solvent. As the compression is increased, the originally spherical microparticle becomes oblate, extending along the radial direction in the x-y plane and contracting along the z-direction. As the compression is increased, the measured radial extension values $\lambda_r = d /do$ are 1.058, 1.116, and 1.217 (Fig. 4b, c, and d, respectively). Here, $d$ and $do$ are the deformed and undeformed diameters of the microparticle, respectively.
Fig. 3. Cross-polarized images of compressed microdisks. The scale bar represents 50 µm. Taken as it is from reference [1].

Fig. 4. Cross-polarized images of a microparticle under various compressions. Compression is increased by 17 µm in each step from (a) to (d). Particle diameter is measured to be (a) 69 µm, (b) 73 µm, (c) 77 µm, and (d) 84 µm. Standard errors can be ±1 µm. Taken as it is from reference [1].
Fig. 5. Schematic illustrations of two possible mesogen orientations in the microparticle: (a) tangential and (b) radial orientations. Taken as it is from reference [1].

Fig. 6. Birefringence of a compressed LCE microparticle. The slow axis of the retardation plate is oriented in the NE-SW direction, at 45° clock-wise from the analyzer (A). The angle denoted on each picture is the rotation angle of the sample stage. Taken as it is from reference [1].
Fig. 7. Single-quadrant model of a microparticle in the initial configuration (a) and the final configuration after deformation (b); and the Green-Lagrange 1st (c) and 2nd (d) principal strain fields at the midplane after the final deformation. The color scale of (b) represents the radial stretching ($\lambda r-1$) of each element, and the color scale of (c) and (d) represents the values of the 1st and 2nd principal strains, respectively. The black lines of (c) and (d) represent the direction of the 1st and 2nd principal strain, respectively; z is the direction of compression, r is the radial direction, and $\theta$ is the circular direction. Taken as it is from reference [1].

The observed patterns in Fig. 3 and Fig. 4 can originate from either a tangential or radial director orientation, as illustrated in Fig. 5. To determine the actual director orientation, we examined the birefringence of the microparticle using a polarized light microscope with a first-order 530 nm full-wave retardation plate, the slow axis of which is 45° to the polarized light. Without any sample in the optical path of polarized white light, the retardation plate induces elliptical polarization of light with wavelength other than the 530 nm as it passes through the
retardation plate. However, green light (530 nm) travels through the retardation plate maintaining its plane polarization and is therefore absorbed by the analyzer. In this case, a bright magenta background is observed [39]. When a birefringent sample with a wavefront ellipsoid parallel to the retardation plate is inserted into the optical pathway, the relative phase retardation of orthogonal wavefronts is changed. If the slow axis of the birefringent sample is parallel to that of the retardation plate, the relative phase retardation increases. As a consequence, the red color is linearly polarized, while the blue and green wavelengths are elliptically polarized and form a hue similar to second order blue. On the other hand, if the slow axis of the birefringent sample is perpendicular to the that of the retardation plate, elliptically polarized green and red wavelengths recombine to form a first order yellow color, while the relative retardation of the orthogonal wavefronts is decreased across the view field and the shorter (blue) wavelengths emerge as linearly polarized light and is absorbed by the analyzer. The birefringence patterns of the compressed microparticle in various orientations are shown in Fig. 6; the slow axis of the retardation plate is oriented in the NE-SW direction. The microparticle at the center of the stage is rotated in 10° increments to show changes in the birefringence pattern. Except for the center part, the interference pattern does not change, implying that the microparticle is axisymmetric. The birefringence pattern shows blue color on the NW and SE quadrants of the particle while yellow on the NE and SW quadrants suggesting that the director in the plane is tangential, as illustrated in Fig. 5a. Previous experimental and numerical studies have shown that the director field in LCE follows the extension direction of LCE sheets and strips [5, 40–42]. In the case of uniaxial extension of a strip, the direction of the positive 1st principal strain aligns with the extension direction, while the directions of the negative 2nd and 3rd principal strains are perpendicular to the extension direction.
However, the strain fields of a compressed sphere can be quite complex. To find the directions of the principal strains of the compressed LCE microparticle, finite element analysis (FEA) using the FEBio package [43] has been carried out. We used 4,000 8-node hexahedral elements (Fig. 7a) and the Mooney- Rivlin (M-R) hyper elastic model that is commonly used to describe the deformation energy of crosslinked elastomers [44]. We note that the M-R model does not include the Frank elastic energy due to the director distortion. We estimate the Frank distortion energy density to be $K/R^2$, where $K$ is the average Frank constant ($\sim 10^{-11}$ N) and $R$ is the radius of the particle ($\sim 30$ μm), and the elastic deformation energy density to be $E\varepsilon^2$, where $E$ is the Young’s modulus ($\sim 105$ Pa), and $\varepsilon$ is the strain ($\sim 0.3$, See Fig. 7c). Therefore, the Frank distortion energy density ($\sim 10^{-2}$ Pa) is negligible to the elastic deformation energy density ($\sim 104$ Pa), and the M-R model is sufficient to describe the strain at the final state of the deformation. In the simulation, the particle is compressed until the radial extension of the outer edge ($\lambda_r - 1 = 0.217$ in Fig. 7b) corresponds to the experimental value in Fig. 3d: $(84-69)/69 \approx 0.217$. We evaluate the principal strains and directions of each node, which are the eigenvalues and eigenvectors of the rotationally invariant Green-Lagrange strain tensor, respectively. The $1^{st}$, $2^{nd}$, and $3^{rd}$ principal strain fields ($E_1$, $E_2$, and $E_2$) are set in the order of their magnitude. Fig. 7c and 7d show $E_1$ and $E_2$, respectively, in the midplane of the compressed microdisk. In fact, the directions of $E_1$ are mostly circular, while those of $E_2$ are mostly radial. All the $3^{rd}$ principal strains ($E_3$) are negative and their directions are in the compression direction (not shown). Most importantly, the magnitudes of $E_1$ are significantly larger than those of $E_2$ at the outer part of the compressed microdisk, implying that extension in the circular direction is the dominant deformation in the outer part of the disk. This is in good agreement with the experimental finding deduced from the birefringence patterns (Fig. 6).
Such a unique tangential director orientation will affect local refractive indices, and develop characteristic birefringence profiles, which could be used as a micro q-plate [45–47], a converging lens [48, 49] or a tunable polarization converter [50]. Fabrication of liquid crystal microlenses, based on an inhomogeneous refractive index profile, involves expensive fabrication procedures and requires clean room facilities [51–53]. Using microfluidics and by sandwiching the microparticles between glass plates an inexpensive solution might be possible, which can be explored. Also, the differing pitch lengths of the helices, due to axisymmetry produced by the compression, may lead to another very interesting application. Cholesteric liquid crystal lasers, with wavelengths being tuned by the helix have been reported previously [54, 55]. The variation in the pitch length of the microparticles, when mixed with a laser dye having a wide emission spectrum, may result in a tunable multi-directional lasing and may generate a range of wavelengths [56].

1.4 Conclusions

I have produced poly-domain LCE microparticles with diameters in the range of 20–85 μm using glass capillary microfluidic devices. Polarized micrographs of birefringence patterns indicate that compressing LCE microparticles induce a characteristic tangential director configuration due to the major extension in the tangential direction, which is confirmed by a three-dimensional finite element analysis.

Acknowledgment

I thank NSF for the materials provided by the New Liquid Crystal Materials Facility (DMR-0606357) and the Kent State University start-up fund for financial support.
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