NEW ELECTRO-OPTICAL APPLICATIONS OF LIQUID CRYSTALS: FROM BEAM STEERING DEVICES AND TUNABLE LENSES TO NEGATIVE REFRACTION AND FIELD-INDUCED DYNAMICS OF COLLOIDS

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by

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CHAPTER 1

Introduction

1.1 Liquid crystals: an amazing class of soft matter materials

Liquid crystals (LCs) are an amazing class of soft matter materials possessing remarkable intrinsic properties [1, 2]. Being discovered in 1888 by Friedrich Reinitzer [3] they attracted a significant scientific interest only in the second half of last century. What makes them so remarkable? The liquid crystalline phases possess anisotropic properties, which can be easily changed under external fields, either mechanical, magnetic or electric. The electric field effect is of a particular interest for numerous practical applications [4]. Nowadays it is rather difficult to imagine our life without LCs as they are at the core of display technologies. Besides the displays applications, LCs found their niche in various electro-optical devices, including beam steerers, optical switches and shutters, tunable lenses, cholesteric mirrors and filters. Other applications include thermal and biological sensors, art and even shampoos [5]. The liquid crystalline order is also pertinent in polymers, emulsions, and biological systems. The list could be endless. Well known is the fact that DNA may form a LC phase even in the case of relatively short complexes [6]; that leads to the speculations on the importance of liquid crystalline phase for the evolution of life. Thus, LCs are really the subject of study for many disciplines. For many decades they are puzzling physicists, chemists, biologists and mathematicians.
LCs are anisotropic fluids usually comprised of elongated organic molecules. This specific anisometric shape of the molecules results in their ordered arrangement leading to an appearance of various liquid crystalline phases (mesophases). For thermotropic LCs, in which the phase transitions occur under temperature changes, the most common observed mesophases are nematic and smectic phases. In the nematic phase the molecules do not possess positional order but have a long-range orientational order; they tend to be parallel to some common axis \([1]\), called the director \(\mathbf{n}\) (a unit vector depicting the average orientation of LC molecules; states \(\mathbf{n}\) and \(-\mathbf{n}\) are equivalent). Deformations of nematic are described by the free energy density as follows \([1, 2]\):

\[
F = \frac{1}{2}K_1(\text{div}\mathbf{n})^2 + \frac{1}{2}K_2(\mathbf{n} \cdot \text{curl}\mathbf{n})^2 + \frac{1}{2}K_3(\mathbf{n} \times \text{curl}\mathbf{n})^2, \tag{1.1}
\]

where \(K_1\), \(K_2\) and \(K_3\) are elastic constants associated with splay, twist and bend deformations, respectively. Optically, nematic is a uniaxial medium with the optic axis along \(\mathbf{n}\). In LC cells \(\mathbf{n}\) can have different orientation of polar angle \(\theta_0\) with respect to the bounding substrates (\(xy\) plane): parallel to the substrates (planar alignment, \(\theta_0 \approx 0\)), perpendicular (homeotropic alignment, \(\theta_0 = 90\)) or tilted \((0 < \theta_0 < 90)\) depending on the alignment set at the LC/bounding substrate interface by the aligning material (buffed polymer film, for example), Fig.1.1 (for the simplicity we assume that the azimuthal angle \(\varphi_0\) is equal to 0). Tilted geometry is important for reducing the switching time of the devices (as discussed in Chapters 3 and 4). At the nematic/surface interfaces the phenomenon called anchoring appears \([1, 2]\), i.e. orientation of the director along one or more directions. Typically anchoring stems from anisotropic character of molecular
interactions such as van der Waals interactions between the LC molecules and those of the aligning material [2]. Strength of anchoring is characterized by the anchoring coefficient $W$, which is equal to the work (per unit area) required to deviate the director from the preferred orientation by a certain angle [2]. Anchoring phenomena are responsible for restoring the director orientation in a cell when the electric field is switched “off”. When the field is switched “on”, anchoring prevents the LC molecules from being completely reoriented. Anchoring phenomena also lead to the director distortions appeared around the particle introduced into a homogeneous director field (as discussed in Chapter 5).

In smectic phase the molecules are arranged in a layered structure with a well-defined interlayer spacing [1] and possess a long-range orientational order as well as a positional order in one direction. While there are numerous types of smectic phase, Smectic A (SmA) and smectic C (SmC) are the most commonly observed. In SmA phase
the director is perpendicular to the layer plane. In SmC the director is tilted within the layer; the tilt value varies depending on the material and temperature.

It is worth to mention an existence of chiral nematic phase (the so-called cholesteric phase), in which the director is continuously twisting to form a helicoidal spiral with a pitch often in the range $0.25 - 10 \mu m$. Cholesteric LCs are capable of reflecting light of certain wavelength. This selective reflection makes them useful for a number of applications ranging from displays [7], to simple consumer products, such as paints and shampoos [5].

The anisometric shape of molecules is responsible not only for the orientational ordering but also results in anisotropic properties of LCs along the director and perpendicular to it. Optical anisotropy of LCs $\Delta n = n_e - n_o$ results in double refraction of light passing through the slab of LC material [4, 8]. Here $n_e$ and $n_o$ are the LC refractive indices for the light propagating perpendicular to $n$ (extraordinary wave) and parallel to $n$ (ordinary wave), respectively. Anisotropy of dielectric constants $\Delta \varepsilon = \varepsilon_|| - \varepsilon_\perp$ leads to the different response of $n$ to the external electric field $E$: if $\Delta \varepsilon > 0$ then the electric field tries to reorient $n$ parallel to itself, $n \parallel E$; in the opposite case $\Delta \varepsilon < 0$ the director would reorient perpendicular to $E$. Here $\varepsilon_||$ and $\varepsilon_\perp$ are the dielectric constants of LC in the directions parallel and perpendicular to $n$. The electrical field-induced director reorientation is very important phenomenon for practical applications. The field-induced reorientation was discovered in 1930s initially for magnetic field [9]. Now, the electrically-controlled nematic cells are at the core of all electro-optical applications.
Fig. 1.2. Sketch of the electrically-controlled director reorientation effect observed in nematic cell placed between crossed polarizers. (a): Field “off” state. (b): Field “on” state. In general, the intensities of the light passed through the system in two cases are not equal: $I_1 \neq I_2$.

Consider a LC cell placed between two crossed polarizers with the LC director $\mathbf{n}$ is in the $yz$ plane initially ($\theta_0 = 0$, Fig.1.1a), Fig.1.2. If the electric field applied along
the $z$ axis exceeds the threshold value $U > U_{th} = \pi \sqrt{\frac{K}{\varepsilon_0 \Delta \varepsilon}}$ [4] then the director $\mathbf{n}$ would reorient from the initial state along the field provided that $\Delta \varepsilon > 0$, Fig. 1.2b. The intensity of the light passed through the system is the function of the angle $\theta$, which director $\mathbf{n}$ makes with the $z$ axis, angle $\phi$, which polarizer makes with the projection of the director on the $xy$ plane, wavelength $\lambda$, extraordinary $n_e$ and ordinary $n_o$ refractive indices of the LC and cell thickness $d$. It can be expressed as follows [4]:

$$I = I_0 \sin^2{2\phi} \sin^2{\left(\frac{\Delta \Phi}{2}\right)},$$

(1.2)

where $\Delta \Phi = \frac{2\pi d}{\lambda} \int_{0}^{d} (n_{\text{eff}}(z) - n_o)dz$ is the phase difference between ordinary and extraordinary waves, $n_{\text{eff}}(z) = \frac{n_en_e}{\sqrt{n_e^2 \sin^2{\theta(z)} + n_c^2 \cos^2{\theta(z)}}}$ is the $z$-dependent refractive index, $I_0$ is the intensity of the incoming light. As the angle $\theta$ changes under the field, then, in general, the intensities of the light passing through the system when the field is “off” and when the field is “on” are not equal, $I_1 \neq I_2$. With different electric field the output intensity varies reaching maximum, minimum and intermediate (grey scale) levels. Thus, a light beam passing through the LC cell experiences a phase shift between the ordinary and extraordinary waves, changes its polarization state, which results in the modulation of the output intensity of the beam behind the output polarizer (analyzer). It is that principle of a phase and intensity modulation, which is at the core of electro-optic applications. Some of them will be discussed in the following Chapters.
1.2 Motivation and objectives of the thesis

Modern electro-optical applications require low cost, fast operation speed and low weight. LCs are providing all these features, but there are problems still unsolved and there are phenomena still unexplored. Technological boom of 20th century led to many discoveries and products including LC displays (LCD) technologies [10]. When I started my PhD, LCDs almost surpassed cathode ray tube TVs in sales. At that time it was realized that there is a moment to look for new phenomena, new materials, new designs and new applications involving LCs, for the applications, which would allow controlling the light not only in the plane of the display, but also in 3D space. The most intriguing areas of the research were adaptive optics, tunable lenses, beam steering devices, development of metamaterials for negative refraction [11], LC-based colloidal dispersions as a novel type of soft matter materials, in which only static interactions were explored and was not much known about the field-induced colloidal dynamics [12], etc. Besides new technologies there was a great demand in improving of the existing devices. For example, LCDs has been known for couple decades, but a relatively slow response time is still an issue. A principle of operation of 90° twisted nematic (TN) cell has been known for almost 50 years [13, 14], but the development of fast polarization rotator with a wide bandwidth is still in progress. Obviously, there are a lot of opportunities for the improvement of the existing LC devices and development the new ones, stemming from the recent discoveries either through the modification of material properties, device designs or both. For example, one can utilize dual-frequency nematics to decrease the switching time of the device. Furthermore, a combination of dual-frequency nematic, a
high pretilt alignment of $n$ at the boundaries and an overdriving switching scheme would improve the response time even more dramatically [15].

All the listed points stipulated the choice of the dissertation topic. In the Dissertation we explored the following directions:

- application of SmA materials for inexpensive birefringent prisms;
- possibilities to improve switching characteristics of TN cells;
- field-induced dynamics of colloidal particles dispersed in LC.

We developed the following devices:

- broadband polarization rotator based on two 45° TNs with opposite handedness;
- LC-based lens electrically tunable from negative to positive regime;

We observed the following phenomena:

- electrically-controlled amphoteric (negative and positive) refraction of light in nematic LC in the certain range of incident angles;
- levitation, selective movement of the particles dispersed in nematic LC towards opposite substrates according to the orientation of satellite defects and bidirectional motion of the colloidal particles controlled by backflow.

1.3 Structure of the thesis

The thesis deals with electro-optical application of LCs. It describes new designs to improve the performance of known devices, such as beam deflectors, polarization rotators and tunable lenses as well as presents relatively new phenomena, such as
electrically-controlled amphoteric refraction of light in nematic LC and electrically-controlled dynamics of colloidal particles immersed in LCs.

The introductory part presents a short overview of LC properties, describes electrically-controlled director reorientation effect, which is at the core of all electro-optical applications of LCs, clarifies the motivation behind the choice of the dissertation topic and explains the objectives of the thesis. In the following four Chapters the results of the conducted research are presented in detail.

Chapter 2 explains the possibility of negative refraction in regular birefringent materials in the certain range of angles of the light incidence and for the first time presents the experimental observation of electrically tunable amphoteric refraction in nematic LC.

Chapter 3 describes applications of SmA materials for passive birefringent prisms useful for digital beam steering and dual-frequency nematics for fast electrically switchable broadband polarization rotators based on TN cells.

Chapter 4 describes applications of dual-frequency nematic for electrically tunable lenses based on hole-patterned electrode structure. The proposed design allows achieving both positive and negative lens utilizing the very same nematic cell.

Chapter 5 presents the results of electric field-induced dynamics of colloidal particles dispersed in nematic LCs. Particles levitation, selective movement toward the opposite substrates, bidirectional translational motion controlled by a backflow are discussed.
Chapter 6 summarizes the results of the research presented in the dissertation and discusses the possibilities for future developments.

1.4 References

5. See, for example, the website: http://www.pantene.com/en-US/product/blondegEINVAL_highlightexpress_dailycolorenhanceshampoo.jspx.
CHAPTER 2

Realization of tunable amphoteric (positive and negative) refraction in nematic liquid crystal

2.1 Introduction

Refraction is an optical effect associated with bending of light passing from one transparent medium to another. The phenomenon is caused by the change of the velocity of a propagating light wave. If two media are isotropic then refraction is described by Snell’s law: \( n_1 \sin \theta_1 = n_2 \sin \theta_2 \), where \( \theta_1 \) and \( \theta_2 \) are the incident and refraction angles of light, respectively, \( n_1 \) and \( n_2 \) are the refractive indices of two media, Fig.2.1. It is obvious, that if \( n_1, n_2 > 0 \) then for \( 0 < \theta_1 < \pi / 2 \) one should expect \( 0 < \theta_2 < \pi / 2 \). In other words the incident and refracted rays are on the opposite sides of the surface normal in this case. Such refraction refers to normal or positive refraction. If incident and the refracted rays are on the same side of the surface normal (if \( \theta_1 > 0 \) then \( \theta_2 < 0 \) ) then we are dealing with the so-called negative refraction [1, 2]. The possibility of negative refraction occurrence in media was theoretically explored by Veselago [3] although some aspects of negative refraction were discussed earlier (see the references in [1, 2]). Veselago predicted that the negative refraction can occur at the interface between two materials when one of them possesses negative values of both electric permittivity \( \varepsilon \) and
magnetic permeability $\mu$ (the so-called left-handed medium or LHM), while another material possesses both positive $\varepsilon$ and $\mu$ (the so-called right-handed medium or RHM). Although LHMs are not found in nature, they can be constructed artificially (the so-called metamaterials) typically by arranging small (sub-wavelength) anisotropic particles of various shapes in space. Why LHMs are so remarkable? Light is an electromagnetic wave with two components, electric $E$ and magnetic $B$, but only electric component effectively interacts with atoms of an ordinary media [1, 2]. It can be seen from the so-called Lorentz law, which expresses the force exerting by electromagnetic field $(E, B)$ on a charge $e$ moving with velocity $v$ as follows: 

$$
F = e(E + \frac{1}{c}v \times B)
$$

[4]. Here $c$ is the speed of light. The term $1/c$ indicates that the force on the electrons in atoms exerted by magnetic field is much weaker than the force due to electric field. LHMs, if created,
would allow both light components effectively couple to the atoms of metamaterial due to resonant enhancement of the light-matter interaction, leading to new remarkable optical applications, such as a “superlens” with sub-wavelength resolution [5, 6]. Other applications of metamaterials may include nanolithography and cloacking [7, 8].

Although LHMs can be constructed for infrared and microwave ranges [1, 2], the current LHMs designs are not yet practical for the visible spectrum. The problem is related to the technological difficulties, as to achieve a negative refraction with low losses in visible part of spectrum, one needs to arrange structural elements (metal-dielectric nanostructures, such as split rings and rods, for example [1, 2]) with the size of less than 100 nm into regular arrays. In the meantime, a lot of the research is directed to analyze whether and how the conventional materials with positive refractive index $n$ can be designed to mimic some features of LHMs [1]. Though, a phenomenon of double refraction of light in birefringent materials was observed a long time ago (in 1669, as stated in [9]), probably a full understanding of possibility of negative refraction occurrence in RHMs was achieved, surprisingly, not a long time ago, in photonic crystals [10] and birefringent solid crystals such as YVO$_4$ [11-14], including experimental proof. The physics behind this is that the directions of wave and energy propagation for the extraordinary mode in birefringent medium do not coincide.

In this Chapter we demonstrate the phenomenon of tunable amphoteric (positive and negative) refraction in LCs [15]. The advantage of LCs over the solid birefringent crystals is that the light propagation can be easily controlled by electric field that changes the optic axis (the director $\mathbf{n}$) orientation. By employing a special cell design that sets a
large (~45°) tilt of \( \mathbf{n} \) at the front interface, we demonstrate that the negative refraction does exist in a uniaxial nematic LC and that it can be switched into the positive refraction regime by an applied electric field.

2.2 Theoretical consideration of negative refraction in nematic liquid crystal

Let us consider a propagation of light in a uniaxial material (nematic LC). In birefringent media in contrast to isotropic media, two linearly orthogonally polarized waves (ordinary and extraordinary) with different phase velocities propagate in a certain direction (the so-called double refraction phenomenon). To solve the problem of light propagation it is useful to build auxiliary surfaces, which allow us to find geometrically wave characteristics in the medium [4, 16]. Consider a planar interface \( z = 0 \), which separates an isotropic medium of a refractive index \( n_i \geq 1 \) occupying the half-space \( z < 0 \), from a nematic LC with the ordinary index \( n_o \) and the extraordinary index \( n_e > n_o \), at \( z > 0 \). The optic axis \( \mathbf{n} \) is in the \( xz \) plane, making an angle \( \alpha \) with the \( z \) axis (normal to the interface between two media), Fig.2.2. Consider a plane wave linearly polarized in the \( xz \) plane entering LC with the angle of incidence in the isotropic medium equal to \( \theta_i \). To find the directions of rays (energy) propagation in LC we build the wave vector surface (\( \mathbf{k} \)-surface) by plotting vectors in all directions from the origin, which lengths are inversely proportional to the phase velocity of a propagating wave. In LC the direction of wave propagation \( \mathbf{k}_i \) does not coincide with direction of energy
Fig. 2.2. Voltage-controlled amphoteric refraction at the isotropic-uniaxial nematic interface. The incident beam is polarized in the plane of figure. The wave vector surfaces are shown as a solid curve for the extraordinary wave and as a dotted circle for the ordinary wave in the nematic LC. In the regime of negative refraction no voltage is applied and the optic axis $\mathbf{n}$ is in the plane of figure making a large angle $\alpha$ with the $z$ axis. The energy flow direction $\mathbf{S}_r$ in the LC is perpendicular to the tangent to the $\mathbf{k}$-surface at point $\mathbf{k}_i$ and thus is different from the direction of $\mathbf{k}_i$. When the voltage is applied, $\mathbf{n}$ reorients along the $y$ axis to establish positive refraction: both the wave vector and energy flow are along the dotted vector.
propagation (Poynting vector) $\mathbf{S}_r$. The dispersion equation for the normalized components of the wave vector found from the Maxwell’s equations reads in the isotropic medium as

$$k_{ix}^2 + k_{iz}^2 = n_i^2, \quad (2.1)$$

and in the LC as

$$\frac{k_{ix}^2}{n_e^2} + \frac{k_{iz}^2}{n_o^2} = 1. \quad (2.2)$$

In the $x'z'$ plane (the $z'$ axis is parallel to the optic axis $n$) the components of transmitted wave vector $\mathbf{k}_t$ are

$$k_{ix} = k_e \cos \alpha + k_o \sin \alpha, \quad k_{iz} = k_e \cos \alpha - k_o \sin \alpha. \quad (2.3)$$

By substituting Eq.(2.3) into Eq.(2.2) one finds that

$$a k_{ix}^2 + b k_{iz}^2 + c k_{ix} k_{iz} = 1, \quad (2.4)$$

where $a = \sin^2 \alpha / n_e^2 + \cos^2 \alpha / n_o^2$, $b = \cos^2 \alpha / n_e^2 + \sin^2 \alpha / n_o^2$, $c = \sin 2\alpha (n_e^{-2} - n_o^{-2})$, and the components of the transmitted wave vector $\mathbf{k}_t$ define the angle $\theta_{t,k} = \tan^{-1} k_{ix} / k_{iz}$, see Ref. [16]. The direction of the refracted time averaged Poynting vector $\mathbf{S}_r$ characterized by the angle $\theta_{r,s} = \tan^{-1}(S_{ix} / S_{iz})$ that $\mathbf{S}_r$ makes with the $z$ axis is perpendicular to the tangent to $\mathbf{k}_t$- surface, and at any point $(k_{ix}, k_{iz})$ it can be found from the equation $\tan \theta_{r,s} = \frac{\partial \phi}{\partial k_{ix}} / \frac{\partial \phi}{\partial k_{iz}}$, where $\phi(k_{ix}, k_{iz}) = a k_{ix}^2 + b k_{iz}^2 + c k_{ix} k_{iz}$, Fig.2.2.

One can find that $\frac{\partial \phi}{\partial k_{ix}} = 2ak_{ix} + ck_{ix}$, $\frac{\partial \phi}{\partial k_{iz}} = 2bk_{iz} + ck_{iz}$. Using the tangential boundary
condition \( k_{ix} = k_{ix} = n_i \sin \theta_i \) at \( z = 0 \), and expressing \( k_{ix} \) through \( k_{ix} \) from the Eq.(2.4) as

\[
k_{ix} = -\frac{ck_{ix}}{2b} + \frac{1}{2b} \sqrt{(c^2 - 4ab)k_{ix}^2 + 4b},
\]

one finds that

\[
\theta_{r,k} = \tan^{-1} \left( \frac{2bn_in_e \sin \theta_i}{2\sqrt{b n_o^2 n_e^2 - n_i^2 \sin^2 \theta_i - cn_on_e \sin \theta_i}} \right), \tag{2.5}
\]

\[
\theta_{r,s} = \tan^{-1} \left( \frac{2n_i \sin \theta_i + cn_on_e \sqrt{b n_o^2 n_e^2 - n_i^2 \sin^2 \theta_i}}{2bn_in_e \sqrt{b n_o^2 n_e^2 - n_i^2 \sin^2 \theta_i}} \right). \tag{2.6}
\]

Similar results were obtained by Luo et al. [14] in a somewhat different way. If \( \theta_i = 0 \), then \( \theta_{r,k} = 0 \); a small increase \( \theta_i > 0 \) makes \( \theta_{r,k} > 0 \), too. The behavior of \( \theta_{r,s} \) is different: for \( \theta_i = 0 \) it can be negative, provided that \( \alpha \neq 0, \pi/2 \), as

\[
\theta_{r,s} (\theta_i = 0) = -\tan^{-1} \left( \frac{n_e^2 - n_o^2}{n_i^2 \sin^2 \alpha + n_e^2 \cos^2 \alpha} \right). \tag{2.7}
\]

In this case the tangential component of the time-averaged Poynting vector changes its sign, i.e., the incident and the refracted rays are on the same side of the surface normal meaning that we arrive into regime of negative refraction. We confirmed these results in the experiment.

### 2.3 Experimental realization of amphoteric refraction in nematic liquid crystal

The experimental setup is shown in Fig.2.3. The He-Ne laser beam (\( \lambda = 633 \text{nm} \)) passes through the polarizer \( P \) to acquire a linear polarization in the \( xz \) plane, and is focused by the 10x microscope objective \( O \) into the middle of the cell filled with nematic mixture E7 (EM Industries, Inc.).
Fig. 2.3. Experimental set-up for observation of tunable amphoteric refraction. The SiO$_x$ coated glass plate is the entry plate for the laser beam. At zero voltage, the director $\mathbf{n}$ is in the $xz$ plane. The electric field $\mathbf{E}$ serves to reorient $\mathbf{n}$ along the $y$ axis.

Objective is necessary to couple an approximately 1mm (in diameter) laser beam into the LC cell. The filter helps controlling the intensity of the laser beam, which is in our case of about 5W/cm$^2$ for the diameter of the focused spot of about 10$\mu m$. This intensity is well below the one that causes an optically-induced nonlinearity of $\mathbf{n}$ [17] and generation of the soliton-like formations, the so-called nematicons [18, 19].

Propagation of the beam is monitored by the CCD camera facing the $xz$ plane of the cell through the microscope objective. The LC cell of thickness $d = 90\mu m$ is assembled using two glass substrates with conductive indium tin oxide (ITO) layers. The substrates are coated with a polyimide PI2555 (Microsystems) film rubbed to align $\mathbf{n}$ uniformly parallel to the $xz$ plane but under the angle $\alpha = (45 \pm 5)^0$ to the $z$ axis. The third glass plate of a crucial importance is an entry glass plate perpendicular to the first
two. This plate imposes a highly tilted, $\alpha = (45 \pm 5)^0$ orientation of $\mathbf{n}$ at the entry interface achieved by an obliquely deposited (at $-5^0$) layer of SiO$_x$ in a vacuum [20-22], thus simultaneously preserving the uniformity of $\mathbf{n}$ throughout the entire cell, Fig.2.3. The LC cell can be rotated around the $y$ axis to control $\theta_r$.

In Fig.2.4 we show the photographs of the electrically tunable amphoteric refraction. When no field is applied the incident beam ($\theta_i = 5.5^0$) is experiencing a negative refraction regime ($\theta_r \approx -4.2^0$), Fig.2.4a. Under electric field $U = 5$ V the refraction switches into the positive regime ($\theta_r \approx 3.8^0$), Fig.2.4b. With the following characteristics for the nematic mixture E7 used in our experiments $n_o = 1.52$, $n_e = 1.74$ (both at 633 nm and room temperature) and $n_i = 1$ (air), $\alpha = 45^0$, one can easily find the range of incident angles, where a negative refraction regime can occur. As clear from Eq.(2.6), there is a critical angle of incidence, $0 < \theta_i < \theta_{cr}$, where $\theta_{cr} = 12.5^0$, below which the refraction is negative. The maximum (negative) angle of refraction

$$\theta_{r,s}^{\max} = -\tan^{-1} \frac{n_e^2 - n_o^2}{2n_o n_e} = -7.7^0 \text{ is achieved when } \theta_i = 0 \text{ and } \alpha = \tan^{-1} \frac{n_e}{n_o} \approx 49^0.$$

When $\mathbf{n}$ is reoriented parallel to the $y$ axis, the refraction regime is positive. The wave vector and the ray vector would propagate at the same angle $\theta_r(\theta_i)$ defined from:

$$n_i \sin \theta_i = n_o \sin \theta_r.$$  \hspace{1cm} (2.7)
Fig. 2.4. Photographs of voltage-controlled amphoteric refraction of a laser beam linearly polarized in the plane of figure on passing through the air into the entry glass plate, \( \theta_i \approx 5.5^0 \), and then into the LC. (a): At zero voltage refraction is negative, \( \theta_r \approx -4.2^0 \); (b): At \( U = 5 \text{ V} \) refraction switches into the positive one, \( \theta_r \approx 3.8^0 \). Corresponding experimental set-ups and arrangements of the director \( \mathbf{n} \) for each regime of refraction are shown schematically in the top row.
Fig. 2.5. Theoretical and experimental data for the electrically-controlled amphoteric refraction in E7. The extraordinary regime dependency $\theta_r, s(\theta)$, Eq.(2.6), is shown by the solid line and the ordinary regime dependency $\theta_r, s(\theta)$, Eq. (2.7), by the dashed line. In the extraordinary case $U = 0$; negative refraction is observed for $0 < \theta < 12.5^0$. In the ordinary case $U = 5 \text{ V}$ (at 1 kHz).
The experimental data for negative and positive refraction ranges and results of calculations are shown in Fig.2.5. The solid line shows the dependence $\theta_{r,s}(\theta_i)$ as calculated from Eq.(2.6). The dashed line shows the dependence of $\theta_r(\theta_i)$ as calculated from Eq. (2.7). Therefore, by applying the electric field to the LC cell and reorienting $n$, one can switch between the negative and positive refraction.

### 2.4 Discussion

The experimental data $\theta_{r,s}(\theta_i)$ and $\theta_r(\theta_i)$ are in a good agreement with the behavior predicted by Eqs. (2.6) and (2.7), respectively, Fig.2.5. Negative refraction occurs for a wide range of the incident angles, $0 < \theta_i < 12.5^\circ$, when there is no field, Figs.2.4a and 2.5. The maximum (negative) bend angle is $\theta_{r,s} = -8^\circ$, as in the theory. By applying the electric field and reorienting $n$ along the $y$ axis, one switches negative refraction to the positive one, Figs.2.4b and 2.5. We deliberately used low voltages ($U = 5$ V) to demonstrate the efficiency of the control; for $d = 90 \mu m$ and $U = 5$ V, the angle between $n$ and the $y$ axis is less than $2^\circ$ within the central portion (of width $\approx 25 \ \mu m$) of the LC cell, as established by numerical simulations of the dielectric response of E7, Fig.2.6. Some deviations of the experimental $\theta_r(\theta_i)$ from Eq. (2.7), noticeable at large $\theta_i$ (Fig.2.5) are caused by the finite width of the region with realigned $n$; as the beam gets defocused in the LC, it propagates in the regions where $n$ deviates
Fig. 2.6. Numerical simulations of director reorientation versus applied voltage in a 90 $\mu$m nematic cell filled with E7. The calculations were performed assuming the following parameters of the nematic material: $K_1 = 11.7$ pN, $K_3 = 19.5$ pN are splay and bend elastic constants of E7, respectively [23]; $\Delta \varepsilon = \varepsilon_{||} - \varepsilon_{\perp} = 19.0 - 5.2 = 13.8$ (EM Industries, Inc.) is the dielectric anisotropy of E7.
from the $y$ axis. Of course, a higher voltage would reorient a larger portion of the nematic cell and thus would refine the ordinary regime, as observed. Note that the intensity of the laser beam was about 5W/cm$^2$; that is far below the critical intensity for the appearance of optically-induced director reorientation, which can be observed at 50W/cm$^2$ [17].

2.5 Conclusions

To conclude, we used one of the simplest possible geometries to prove the very existence of negative refraction and electrically-controlled amphoteric refraction in LCs that can be used in a variety of applications, e.g., beam steering. In our cells with a large $d$, the characteristic time of director reorientation was tens of seconds. However, there is no principal obstacle to speed the response up by reducing $d$, using dual-frequency materials or higher voltages. The biggest advantage of LCs is in the flexibility and controllability of the optical axis $\mathbf{n}$. Even higher degree of flexibility is expected when the LC is used as one of the components in heterogeneous materials, such as photonic crystals [24-26], in which one might expect to expand beyond the regime of negative refraction in the positive index anisotropic material presented in this Chapter.

2.6 References


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CHAPTER 3

Development of liquid crystal-based optical elements for digital beam steering:

smectic A-filled birefringent prisms and broadband polarization rotators

3.1 Introduction

Beam steering devices are in great demand in free space laser communications, optical fiber communications, optical switches, scanners [1-5], etc. In contrast to inertial mechanical devices, such as mirrors, LCs promise an inertia-free operation in non-mechanical beam steerers and deflectors. LCs possess low size and weight, low-cost fabrication [1, 6], fast switching [7] and low operating voltage. The most popular LC-based beam steering devices are based on diffractive and prismatic designs. Diffractive LC devices can provide a continuous steering and are known at least since 1974, when Borel et al. described a binary rectangular LC diffraction grating [8]. This approach has been expanded by an optical-array beam steerers [1, 9-11], polymer-dispersed LC gratings [12], ferroelectric LC gratings [13], voltage-controlled cholesteric LC gratings capable of both Raman-Nath and Bragg diffraction [14-16], etc.

Prism-based digital beam deflectors (DBDs) provide a discrete (digital) steering angles; one of the most effective designs employs a cascade of elementary stages consisting of pairs of an active polarization switch and a passive prismatic deflector [17-
The advantage of such a decoupled design is that it allows one to separate the issue of the short response time determined by a polarization switch and the angular range of deflection determined by the geometry and optical properties of the deflector. For example, such a pair can be comprised of an electrically switchable 90° twisted nematic (TN) cell followed by a passive birefringent prism, Fig.3.1. TN cell rotates the polarization of incident light by $\pi/2$ (field “off” regime) or leaves the polarization intact under applied voltage (field “on” regime). Depending on the polarization of the incoming light, a birefringent prism with different ordinary ($n_o$) and extraordinary ($n_e$) refractive indices and dihedral angle $\alpha$ separates the beam into two channels with different angles of light propagation due to the phenomenon of light refraction briefly discussed in Chapter 2:

$$\theta_e = \sin^{-1}(n_e \sin \alpha) - \alpha,$$

$$\theta_o = \sin^{-1}(n_o \sin \alpha) - \alpha.
\tag{3.1}$$

As clear from the Fig.3.1, if the smectic A (SmA) prism is used, the optic axis (and thus the preferred orientation of the SmA molecules) should be aligned along the edge of the wedge. In this geometry the director field is uniform everywhere. The decoupled pair of a rotator and deflector has no moving parts and can be cascaded into N stages, making $2^N$ addressable beam directions [17-25].

While application of LCs for electrically switchable components is a well-developed field [26], the use of LCs in the passive prismatic elements is less studied despite their apparent advantages, such as structural flexibility and low-cost fabrication. One of the reasons for such a neglect is that a LC-based prism with a substantial dihedral
Fig. 3.1. The scheme of a beam deflecting pair, comprising a switchable polarization rotator (90° TN cell) and a passive prism filled with SmA. (a): In “off” state the TN cell rotates the polarization state of the incoming light by 90°. The light gets deflected by the birefringent prism by the angle $\theta_e = \sin^{-1}(n_e \sin \alpha) - \alpha$. (b): In “on” state (TN cell is under applied field) the polarization state of the incoming light remains intact (parallel to the plane of the figure). The light gets deflected by the birefringent prism by the angle $\theta_o = \sin^{-1}(n_o \sin \alpha) - \alpha$. Here $n_e$ and $n_o$ are the ordinary and extraordinary refractive indices of SmA, respectively.

Angle $\alpha$ (needed for the substantial angle of beam deflection) and a substantial aperture $A$ should be relatively thick, up to $h = A \tan \alpha$. Thickness increase leads to the huge losses caused by light scattering at director thermal fluctuations [27, 28] limiting the applicability of the nematic LCs.
In this Chapter we describe application of SmA for passive optical elements [29] and dual-frequency nematic for fast broadband polarization rotators based on TN cells [30]. SmA prisms have rather high birefringence and can be constructed in a variety of shapes, including single prisms and prismatic blazed gratings of different angles and profiles. The advantage of the SmA materials over nematic LCs is that director fluctuations are suppressed by the layered smectic structure. We address the challenges of uniform alignment of SmA, such as elimination of focal conic domains (FCDs). Fast rotation of the incident light polarization can be achieved by an electrically switched TN cell incorporating dual-frequency nematic and high pretilt alignment set by oblique SiOₓ deposition [7, 31, 32].

3.2 Development of SmA-based birefringent prisms

3.2.1 Alignment of SmA: focal conic domains elimination

In SmA the molecules are arranged in a periodic stack of layers with the director \( \mathbf{n} \) being perpendicular to the layers; the states \( \mathbf{n} \) and \( -\mathbf{n} \) are identical. Inside the layer the molecular centers of gravity show no long-range order, thus each layer is a two-dimensional fluid. Positional order along \( \mathbf{n} \) significantly reduces thermal director fluctuations and light losses caused by scattering [27, 28]. However, the layered structure introduces another possible source of light scattering, namely, static director distortions, such as undulations, and appearance of defects, such as FCDs [33-36]. The problem is especially pronounced for the polar cyanobiphenyl materials, in which the molecules within a smectic layer usually form partially overlapped pairs with oppositely oriented
dipole moments. The thickness of smectic layers in these materials is about 1.4-1.6 of the length of an individual molecule and can vary significantly with temperature. Temperature changes may result in the changes of layers thickness and, in turn, in director distortions. These FCDs can be stabilized by a mechanical impurity in the bulk or at the surface of the cell [33]. In principle, a magnetic field can be used to align the SmA sample uniformly [34, 35]. Let us estimate the field needed to align SmA uniformly.

We consider the simplest type of FCD, the so-called toric FCD that can be stabilized, for example, by a foreign particle in SmA aligned in a bookshelf geometry in a cell with planar (parallel to the substrates) alignment of the director [37, 38]. For the sake of simplicity let us assume that a foreign particle is a disk-like with tangential boundary conditions on its surface. A toric FCD is formed by a pair of linear defects, a circular defect line and the straight line passing through the center. The smectic layers are wrapped around the pair preserving the thickness of smectic layers, Fig.3.2. A toric FCD is a particular case of a general FCD comprised of the ellipse and hyperbola [33]. Note, that outside the FCD the molecules are oriented uniformly along the director; thus, all director distortions are concentrated within a defect with the radius $a$ of the defect circle. Suppose, that $a$ is much smaller than the cell thickness. The elastic energy of a toric FCD can be written as follows [39]:

$$F_{el} = 2\pi^2 aK \left[ \ln \left( \frac{2a}{r_c} \right) - 2 - \frac{\vec{K}}{K} \right] + F_c,$$

where $K$ is the splay elastic constant, $\vec{K}$ is the saddle-splay constant, $r_c$ is the core radius of the circular defect, and $F_c$ is the core energy of the circle and the straight line.
Fig. 3.2. Schematic view of a toric FCD. The SmA layers are perpendicular to the substrates and folding within the FCD. The director changes the direction on 90° from the tangential orientation outside the FCD to the vertical within the FCD to satisfy the boundary conditions at the disk-like foreign particle located in the SmA bulk.

FCD would be stable if its elastic energy $F_{el}$ is smaller than the anchoring energy difference between the FCD-free (uniform) state and the FCD state:

$$
\Delta F_i = 2\pi a^2 W,
$$

(3.3)

where $W$ is the (polar) surface anchoring coefficient at the SmA-particle interface. In SmA, $W \approx \left(10^{-3} - 10^{-2}\right) J/m^2$ is much higher than the corresponding value in the nematic phase, $W \approx \left(10^{-5} - 10^{-4}\right) J/m^2$ [40].
If the anisotropy $\chi_a = \chi_\parallel - \chi_\perp > 0$ of SmA diamagnetic susceptibility is positive, then the applied magnetic field $\mathbf{B}$ in the direction of the desired orientation of molecules should reduce the size of FCD, as the molecules inside the domain should reorient along $\mathbf{B}$, Fig.3.2. The energy gain from such a reorientation is $\Delta F_B = 2 \int \frac{1}{2} \mu_0^{-1} \chi_a B^2 \sin^2 \theta dV$, where $\mu_0 = 4\pi \cdot 10^{-7}$ N/A$^2$ is the permeability of free space, $\theta$ is the angle between the director and $\mathbf{B}$, the volume element is $dV = r(a - r \sin \theta) \sin \theta \theta d\theta d\varphi dr$; $r$ and $r - a / \sin \theta$ are the principal radii of curvature of SmA layers within the toric FCD, $r$ varies in the range from 0 to $a / \sin \theta$; $0 \leq \theta \leq \pi / 2$; $0 \leq \varphi < 2\pi$. Integration yields

$$\Delta F_B = \frac{1}{3} \pi \mu_0^{-1} \chi_a B^2 a^3. \quad (3.4)$$

The stability of the FCD is determined by the energy difference between the FCD state and the uniform state, comprised of the elastic, surface anchoring, and diamagnetic contributions, $\Delta F = \Delta F_s - F_{el} - \Delta F_B$:

$$\Delta F = 2\pi a^2 W - 2\pi^2 aK \left[ \ln \frac{2a}{r_c} - 2 - \frac{K}{K} \right] - F_c - \frac{1}{3} \pi \mu_0^{-1} \chi_a B^2 a^3. \quad (3.5)$$

Figure 3.3 shows the function $\Delta F(B)$ for four different sizes of FCDs of radius $a = 0.5, 1, 2, 5 \mu m$ calculated for the following typical values of LC parameters [28, 40]: $K = 10^{-11}$ N, $K = 0$, $W = 5 \cdot 10^{-3}$ J/m$^2$, $r_c = 10$ nm, $F_c = 0$ ($r_c$ is chosen to adsorb the core energy into the elastic energy of layers distortions [27]), and $\chi_a = 10^{-5}$. The plot demonstrates that for each value of $a$ there is a critical value of the field, for which $\Delta F$ becomes negative, i.e., the uniform state becomes energetically preferred over the FCD.
state. The higher the field, the smaller is the size of the FCDs that can be transformed into the uniform state. However, to reduce the size of FCD to a practical sub-wavelength value, say, $a = 0.5 - 1 \, \mu m$, one needs to apply huge magnetic fields of the order of tens and hundreds of Tesla, Fig.3.3. The defect-free alignment can be achieved by applying the magnetic field while the material is in the nematic phase and then slow cooling it down to the SmA phase, as the surface anchoring in the nematic phase is much weaker that in the SmA phase. The magnetic field needed to realign the director around the foreign inclusion in the nematic phase can be determined from the condition that the magnetic coherence length $\xi = \frac{1}{B} \sqrt{\frac{\mu_0 K}{\chi_a}}$ is smaller than the anchoring extrapolation length $l = \frac{K}{W}$:

$$B_c = W \sqrt{\frac{\mu_0}{K \chi_a}}. \quad (3.6)$$

For example, the field $B = 1 \, T$ would be sufficient to suppress the director distortions around a particle with $W = 10^{-5} \, J/m^2$; $B = 10 \, T$ would be needed if $W = 10^{-4} \, J/m^2$, etc. Therefore, a uniform alignment by magnetic field can be achieved easier in the nematic phase than in the SmA phase. These considerations suggest that in order to minimize light losses caused by a scattering in a thick LC sample, one should search for the SmA material composed of nonpolar molecules, in which: a) the molecules within a layer do not form pairs so the layer’s thickness does not change much with temperature and b) in which there is a nematic phase, in addition to the SmA phase.
Fig. 3.3. Free energy difference between the uniform state of a SmA slab with an incorporated foreign particle-disk and the state with the toric FCD, as the function of the magnetic field applied to align the director uniformly. Different curves corresponding to the radius of the circular FCD base equal to 0.5, 1, 2, 5 µm.

3.2.2 Defect-free alignment of dialkylazoxybenzene SmA materials

The requirements to the defect-free SmA are met by low-molecular weight materials belonging to the class of 4,4'-n-dialkylazoxybenzenes [20, 21], Fig.3.4a. We used n=5,6,7,8 homologues of 4,4'-n-dialkylazoxybenzene (Sigma-Aldrich) to prepare mixtures with a broad temperature range of the SmA phase. All components were purified to decrease the contents of undesired dopants and foreign particles. The purification process was as follows: first we dissolved the compound in methanol (99.93%, Sigma-Aldrich) in proportion 1 gram of LC in 100 ml of methanol. Then the
solution was cooled down to separate the material from the methanol. The precipitated crystals were filtered and dried out. The purified compounds were mixed in various proportions to get the appropriate phase sequence and a good alignment. The temperature range of SmA phase can be expanded to about 30 degrees in eutectic mixtures. The mixture of $n=6$ and $n=8$ homologues in proportion 1:1 showed the lowest light losses and a rather wide temperature range of SmA phase, Fig.3.4b. These mixtures were used as a SmA filler for passive prismatic deflectors.

3.2.3 SmA-based macroprisms

We designed the prismatic cell filled with SmA material as shown in Fig.3.1. Two glass plates with rubbed polyimide layers (PI2555, Microsystems) formed a wedge cell, which was filled with the SmA blend of 4,4’-dihexylazoxybenzene and 4,4’-dioctylazoxybenzene in proportion 1:1. The assembled cell was heated to the isotropic state and slowly cooled down to the room temperature with the temperature rate $5 \times 10^{-4}$ K/s in 1.2 Tesla magnetic field. The measured parameters of the designed birefringent prism were as following:

- wedge angle: $\alpha = 9.2^0 \pm 0.1^0$; 
- refractive indices of the SmA mixture (at $\lambda = 633$ nm and temperature 22$^0$C): $n_e = 1.72 \pm 0.01$, $n_o = 1.53 \pm 0.01$, $\Delta n = 0.19$; 
- angles of deflection: for the light polarized parallel to the LC director (extraordinary wave or $e$-wave) $\theta_e = 6.7^0$, for the light polarized perpendicular to the LC director (ordinary wave or $o$-wave) $\theta_o = 4.9^0$. 

Fig. 3.4. (a): Molecular structure of 4,4’-n-dialkylazoxybenzene. (b): Phase diagram of the binary mixture of n=6 and n=8 homologues of 4,4’-n-dialkylazoxybenzene.

The designed wedge cell showed rather good uniform alignment of SmA material with a small amount of residual isolated FCDs (Fig. 3.5a). For the comparison, the similar wedge filled with 4-octyl-4’-cyanobiphenyl (8CB, EM Industries, Inc.) showed a significant amount of FCDs (Fig. 3.5b) causing a noticeable light scattering. In Fig. 3.6 we show the transmission of the extraordinary and ordinary waves through the wedge filled with the blend of 4,4’-dihexylazoxybenzene and 4,4’-dioctylazoxybenzene in proportion 1:1 in SmA (t=25°C) and nematic phases (t=48°C) at the wavelength $\lambda = 633$ nm. The photodetector was placed at the distance 22 cm from the sample. The diameter of the probing laser beam was about 2 mm. The data are normalized by the incident light intensity $I_0$. Figure 3.6 clearly demonstrates that SmA phase is much more transparent.
than the nematic phase due to reduction of light scattering at the director fluctuations. The transmission of SmA phase remains above 70% for both ordinary and extraordinary light components even when the LC layer becomes thicker than 1 mm (note, no antireflection coatings were used). The variations of light transmission with thickness observed in the plots in Fig.3.6 for SmA are caused by the residual amount of FCDs observed in the wedge-shaped cell used in the measurements. The photographs of the beam passed through the wedge are shown in Fig.3.7. As the SmA mixture has a positive birefringence $\Delta n = n_e - n_o > 0$, the e-wave will deflect more than the o-wave, Figs.3.7a-c.

3.2.4 Polymer array of prisms filled with SmA material

The assembling of the SmA-filled wedges for the wide-aperture incident beam requires large quantities of LC material. Besides, the light transmission through the wedge significantly drops if the thickness of the thick part of the wedge exceeds 2 mm. A more practical approach for wide aperture elements might be to replace a single birefringent prism with an array of smaller prisms, at the expense of some decrease in light transmission efficiency caused by light diffraction, destructive interference and non-ideal prism profile. Hirabayashi et al. reported on quartz microprisms filled with the nematic material, which can deflect closely spaced micro-optical beams individually to any position with a high transmittance, high deflection angle and low voltage [41]. Here we describe a passive element: an array of prisms filled with a SmA material.
Fig. 3.5. Polarization microscope textures of the wedge cells filled with the SmA material after alignment in 1.2 Tesla magnetic field. Thickness of both cells in the center of pictures $\approx 200\mu m$, wedge angle $\alpha = 9.2^\circ$. (a): The wedge filled with the SmA blend of 4,4'-dihexylazoxybenzene and 4,4'-dioctylazoxybenzene in proportion 1:1. Isolated FCDs are observed. (b): The wedge filled with 8CB SmA material. SmA alignment is affected by the bundles of FCDs.
Fig. 3.6. Transmission of the light (e- and o-waves) passed through the birefringent wedge (α = 9.2°) filled with the SmA blend of 4,4’-dihexylazoxybenzene and 4,4’-dioctylazoxybenzene in proportion 1:1 at λ = 633 nm. The data are taken when the mixture is in SmA (t=25°C) and nematic (N, t=48°C) phase.
Fig. 3.7. Deflection of the light passed through the birefringent wedge ($\alpha \approx 9.2^\circ$) filled with the SmA blend of 4,4'-dihexylazoxybenzene and 4,4'-dioctylazoxybenzene in proportion 1:1. (a): Position of the non-deflected beam on the screen. (b): Position of the deflected ordinary beam. (c): Position of the deflected extraordinary beam.

Right angle prisms may be molded in a sheet of polymer material with a different cut angle $\alpha$ and period $d$, Fig.3.8. We used the array of prisms formed in an acrylic films of optical quality with the refractive index $n_a=1.49$ (at $\lambda=589$ nm) with $\alpha=30^\circ$ and $d=1$ mm (Fresnel Technologies, Inc.). The acrylic array of prisms was attached to the glass substrate coated with rubbed polyimide PI2555 to align the director $\mathbf{n}$ along the prism edge. Then the assembled cell was filled with the 1:1 (by weight) mixture of 4,4'-dihexylazoxybenzene and 4,4'-dioctylazoxybenzene. After magnetic field alignment, we measured the light transmission and deflection angles of the cell at $\lambda=633$ nm for normal incidence at the room temperature. We considered the zero-order diffracted beam; the diameter of the incident light beam was 5 mm; the cell was illuminated from the glass
Fig. 3.8. Sketch of the array of polymer prisms filled with the SmA material.

plate side. The results were as follows:

- for the $e$-wave, the normalized light transmission was ~75.4% and the deflection angle $\theta_e=7.20^\circ \pm 0.20^\circ$;

- for the $o$-wave, the normalized light transmission was ~80.5% and the deflection angle $\theta_o=0.60^\circ \pm 0.20^\circ$.

The performance of the assembled cell is very similar to Rochon prisms, but due to a slight mismatch between the $n_a$ and $n_o$ of the SmA the $o$-wave is slightly deflected. We checked the temperature dependence of the deflection characteristics of SmA-filled microprisms. The temperature-induced changes in the deflection angles were relatively small, about 0.02$^\circ$ per 1 $^\circ$C for the extraordinary and less than that for the ordinary wave. Thus, with the temperature increase from 15$^\circ$C to 40$^\circ$C the deflection angles changed from 7.14$^\circ$ to 6.68$^\circ$ for the $e$-beam and from 0.61$^\circ$ to 0.56$^\circ$ for the $o$-beam.
The light propagation through the array of prisms is affected by the geometry of the prisms which is far from the ideal triangular profile. In Fig.3.9 we show the fluorescence confocal polarization microscopy (FCPM) [42] images of the prisms filled with the mixture of Cargille™ refractive index fluid \( n_{dn}^{imm\, oil} = 1.52 \) and Nile Red (Aldrich) fluorescent dye (0.01\% by weight). FCPM technique allows one not only obtain the images in the \( xy \) plane but also resolve the details along the \( z \) axis. It also allows to decipher the orientational order of LC using the property of an anizometric dye to orient itself in anisotropic medium [42]. As one can see the prisms profile is not regular, which is the reason for a difference (of about 0.2°) between the measured and theoretically calculated values of the deflection angles.

The array of prisms may find their applications as a passive optical interconnect, for example, in fiber optics, where multiple beams of small aperture are involved.

3.3 Fast achromatic polarization rotator based on 45° TN cells

3.3.1 Brief overview of polarization rotator concepts

As we already discussed, electrically tunable polarization rotators are of significant interest for beam steering devices. They are also of a prime importance in displays applications, spatial light modulators, optical interconnects, etc. The simplest tunable polarization rotator can be made of electrically-controlled nematic cell capable of
Fig. 3.9. Fluorescence confocal polarization microscopy study of the polymer prisms array. (a): Apex of one acrylic prism of the array. (b): Base of the acrylic prism.

providing a half-wave phase retardation. However, this rotator can operate only in a narrow spectral band where the half-wave condition is satisfied. To avoid a wavelength dependence, a multiple element approaches were developed. For example, two identical half-wave plates oriented in a certain way to the polarization state of the incident light were used for the achromatic rotation of the linearly polarized light in the range 450 – 700 nm [43, 44]. In the three-element design, the ferroelectric LC half-wave switchable plate [45] and a thin 90° TN cell [46, 47] were placed between two passive birefringent plates; the resulting light transmittance was high $T \geq 0.98$ in the visible range 400 – 700 nm. The achromatic polarization switch with a nematic cell sandwiched between two
identical 135° TN cells was designed and showed good performance for visible and near infrared spectral ranges [48, 49].

A TN cell alone can provide a tunable achromatic polarization rotation. In general, in a TN cell the LC director orientation at the opposite substrates makes a certain angle called a twist angle [50, 51]. In the bulk \( n \) is continuously rotating to satisfy the boundary conditions. The direction of twist can be also different so the director can form a right- or left-handed helix. An ‘ideal’ achromatic rotation for a broad optical range with a high contrast may be achieved when all the parameters such as TN cell twist angle \( \Theta \), thickness \( d \), light wavelength \( \lambda \) and the nematic birefringence \( \Delta n \) satisfy the Mauguin condition [52-54]:

\[
d >> \frac{\lambda \Theta}{\pi \Delta n}. \tag{3.7}
\]

In this case the transmission of a TN cell placed between polarizer oriented parallel to the director set at the entrance plate and an analyzer oriented perpendicularly to the director at the exit plate can be calculated as follows [54]:

\[
T = \frac{\sin^2(\Theta \sqrt{1+u^2})}{1+u^2}, \tag{3.8}
\]

where \( u = \pi d \Delta n / \lambda \Theta \). According to the Eq. (3.8) the transmittance has a non-monotinous behavior and goes through the series of decreasing maxima and zeros.

A relatively thick TN cell rotates the linearly polarized light achromatically; the electric field vector of a light wave in this case follows the twisted director structure of the TN cell. Unfortunately, a large \( d \) needed to satisfy this condition causes a long relaxation time of the nematic device:
\[ \tau_{\text{off}} \approx \frac{\gamma_1 d^2}{\pi^2 K}, \]  

(3.9)

where \( \gamma_1 \) is the rotational viscosity and \( K \) is the elastic constant of nematic material [26].

A thick TN cell (\( d \approx 40 \mu m \)) was tested as a polarization switch in infrared at \( \lambda = 10.6 \mu m \) [55]. It was found that \( \tau_{\text{off}} > 0.5 \text{ s} \) in a qualitative agreement with Eq.(3.9) for the typical nematic material with \( \gamma_1 \approx 0.1 \text{ kg \cdot m}^{-1} \cdot \text{s}^{-1} \) and \( K \approx 10^{-11} N \).

Recently it was shown that by using dual-frequency nematic and a high pretilt alignment of the director near the boundaries one can significantly reduce a switching time of rather thick nematic cells [7, 56, 57]. The distinctive feature of a dual-frequency nematic is a frequency dependent dielectric anisotropy \( \Delta \varepsilon(f) = \varepsilon_\parallel(f) - \varepsilon_\perp(f) \), which is positive below some critical frequency \( f_c \) and negative at \( f > f_c \); here \( \varepsilon_\parallel \) and \( \varepsilon_\perp \) are the dielectric permittivities parallel and perpendicular to \( n \). The driving voltage aligns the director parallel to the field \( E \) when \( f < f_c \) and perpendicular to it when \( f > f_c \). The dual-frequency nematic allows one to reduce the relaxation time because the direct and reversed transitions between “on” and “off” states are controlled by the driving voltage of a proper amplitude and frequency. In this case the response times for both cases (switching “on” and “off”) can be written as follows [26]:

\[ \tau_{\text{on/off}} \approx \frac{\gamma_1 d^2}{-\pi^2 K + \varepsilon_0 |\Delta \varepsilon(f)| U^2}, \]  

(3.10)

where \( \varepsilon_0 \) is the permittivity of free space, \( U \) is the applied voltage (RMS value) [26].

Utilizing this approach, an achromatic linear polarization rotator in visible-near infrared...
range \(0.4 \mu m \leq \lambda \leq 2.7 \mu m\) based on 45 \(\mu m\) 90° TN cell with a fast response time of 10 ms and high optical transmission \(T \geq 0.9\) was demonstrated [30]. In the next section we discuss the approach utilizing two 45° TN cells filled with dual-frequency nematic [30].

3.3.2 Design of fast achromatic polarization rotator based on two 45° TN cells filled with dual-frequency nematic

Consider the experimental set-up schematically shown in Fig.3.10. A linearly polarized light beam (the polarization state is set by the input polarizer parallel to the \(y\) axis) propagating along the \(z\) axis is consecutively passing through the right-handed (TN1) and left-handed (TN2) 45° TN cells. The easy axes at the incident and output substrates of the cells (shown by the arrows) are arranged in such a way that \(n||y\) at the input substrates and makes +45° and -45° at the output substrates of TN1 and TN2, respectively. The cells were assembled from ITO glass substrates and filled with the dual-frequency material MLC-2048 \(\left(f_c=12\ kHz\ at\ 20^\circ C\right)\). The thickness of both cells was 20 \(\mu m\); the orientation of \(n\) was set by obliquely deposited SiO\(_x\) that provides a high pretilt alignment (about 45° for MLC-2048) of \(n\) at the boundaries. The right- and left-handed twists in TNs were obtained by a proper arrangement of the glass substrates with SiO\(_x\) layers; no chiral dopants were used. A high pretilt alignment of \(n\) maximizes the initial reorienting torque exerting on the director by electric field \(E\), which is proportional to \(\sin 2\theta\). Here \(\theta\) is the angle between \(n\) and \(E\). The output polarizer (analyzer) can be oriented parallel to the projection of \(n\) set at the output substrate of the TN2 onto the \(xy\)
plane or perpendicular to it. The principle of operation of a pair of the TNs is as follows: if the low-frequency ($f=1$ kHz) voltage is applied to TN1, then the LC director reorients along the field (homeotropic state) so the TN1 does not change the polarization state of the passing light. The polarization state of the light passing through the TN2, which is in a twisted (field “off”) state, is rotated by $-45^0$ counterclockwise. If TN2 is switched “on” while TN1 is “off”, then the polarization state of the incident light is rotated by $+45^0$
clockwise. As a result, depending on states of TN1 and TN2, the output light may have either of two mutually orthogonal linear states of polarization.

The obvious drawback of the proposed scheme is an increased number of the driving channels (two instead of one used, for example, for single 90° TN). However, the advantage of this approach is that 45° TN cells can be approximately 2 times thinner than 90° TN cell because $d \propto \Theta$, see Eq.(3.7). Decreasing the thickness by a factor of 2 makes the cell faster by a factor of 4, see Eq.(3.10). We present the response time measurements in the following Section.

### 3.3.3 Response time measurements

We measured the switching characteristics of a pair of 20 µm thick 45° TN cells sandwiched between polarizers at $\lambda = 633 \text{ nm}$, Fig.3.10. The input polarizer was oriented parallel to the $y$ axis, while the axis of the analyzer was oriented parallel to the projection of $\mathbf{n}$ set at the output substrate of TN2 onto the $xy$ plane. To switch the cells we employed an overdriving scheme when the applied signal is a sequence of high-amplitude pulses (switching voltages $U^{\text{switch}}$) followed by low-amplitude pulses (holding voltages $U^{\text{hold}}$) at frequencies of 50 kHz and 1 kHz [7]. In Fig.3.11 we show the response time measurements of a pair of two 45° TNs. First, TN1 is under low-frequency holding voltages $U^{\text{hold}}_{1 \text{ kHz}} = 50 \text{ V}$, while TN2 is under high-frequency holding voltages $U^{\text{hold}}_{50 \text{ kHz}} = 5 \text{ V}$. This situation is sketched in Fig.3.11a: for TN1 the LC director is in a homeotropic state while for TN2 it is in a twisted state. Then, the signals are reversed: TN1 is under high-
Fig. 3.11. Response time measurements of the pair of 20 \( \mu m \) thick 45° TN cells at \( \lambda = 633 \text{ nm} \). (a, b): Schemes of two possible states of TN cells, homeotropic and twisted, alternately induced by low-frequency (1 kHz) and high-frequency (50 kHz) signals, respectively. (c-d): Oscillograms of cells response (upper trace) together with applied signals to the TN1 (middle trace) and to the TN2 (lower trace). The applied signals are a sequence of high amplitude switching pulses (\( U_{\text{switch}}^{\text{1 kHz}} = 74 \text{ V}, U_{\text{switch}}^{\text{50 kHz}} = 66 \text{ V} \)) and holding voltages (\( U_{\text{hold}}^{\text{1 kHz}} = 50 \text{ V}, U_{\text{hold}}^{\text{50 kHz}} = 5 \text{ V} \)) at two frequencies: 1 kHz and 50 kHz. The response time of each cell is about 2 ms (d, e).
frequency signal $U_{\text{hold}}^{50 \text{ kHz}} = 5 \text{ V}$, while TN2 is under low-frequency signal $U_{\text{1 kHz}}^{\text{hold}} = 50 \text{ V}$.

This situation is sketched in Fig. 3.11b: for TN1 the LC director is in a twisted state while for TN2 it is in a homeotropic state. Thus, both TN1 and TN2 are alternately switched between homeotropic and twisted states so the optical signal is changing from maximum to minimum as the linear polarization of light is switched between two orthogonal orientations, Fig.3.11c. High-frequency holding voltages ($U_{\text{hold}}^{50 \text{ kHz}}$) are necessary to keep $\mathbf{n}$ in the $xy$ plane as SiO$_x$ alignment layer induces a tilted orientation of $\mathbf{n}$. Fast transition from a twisted state to homeotropic was achieved by a low-frequency switching voltages $U_{\text{switch}}^{1 \text{ kHz}} = 74 \text{ V}$ (Fig.3.11d) and from a homeotropic state to twisted by a high-frequency switching voltages $U_{\text{switch}}^{50 \text{ kHz}} = 66 \text{ V}$. The “on” and “off” switching time of both TN cells is about 2 ms, which is in a qualitatively agreement with Eq.(3.10) and two orders of magnitude faster than for the 20 $\mu$m thick cell switched in the conventional mode, according to Eq.(3.9). The fast response, however, is accompanied by a slow drift in the transmitted intensity (less than 5%), which might be related to the backflow effect (flow of the material induced by the reorientation of $\mathbf{n}$). In these measurements the output polarizer was oriented parallel to $\mathbf{n}$ at the output substrate but oscillograms were not changed much when the output polarizer was rotated by $90^0$. Also, oscillograms were not changed much when two other wavelengths were used in response time measurements ($\lambda = 1064 \text{ nm}$ and $\lambda = 1550 \text{ nm}$) utilizing the same frequencies and amplitudes of the applied signals.
3.3.4 Spectral characteristics of 45° TNs

To determine the transmission spectra we placed the system of two 45° TNs sandwiched between polarizers in the sample compartment of the infrared spectrometer (Spectrum One NTS, PerkinElmer). $U_{1\,kHz}^{\text{hold}} = 50$ V was applied to TN1 and $U_{50\,kHz}^{\text{hold}} = 5$ V to TN2 as sketched in Fig.3.11a. Figure 3.12 shows the normalized transmission spectra of the system versus wavelength for two orientations of the analyzer: parallel to the projection of $\mathbf{n}$ set at the output substrate of TN2 onto the $xy$ plane (squares) and perpendicular to it (triangles). The transmission was also calculated for both cases (solid and dash lines, respectively) by using the Eq.(3.8) and assuming that TN1 is in a perfect homeotropic state ($\mathbf{n}$ perpendicular to the boundaries), TN2 is in a perfect twisted state (no tilting of $\mathbf{n}$ near the boundaries) and taking into account the dispersion of MLC-2048.

To measure the optical dispersion of MLC-2048 material we used the wedge cells formed by two glass substrates with polyimide (PI2555) rubbed parallel to the wedges edge. The light passing through the wedge cell will deviate from the initial direction according to Eq.(3.1); the deflection angle will be different for the o- and e-waves. Knowing the dihedral angle $\alpha$ and measure the deflection angles $\theta_e, \theta_o$, one can find the refractive indices of the material for different wavelengths at given temperature. In the experiment we used three available wavelengths: 633 nm, 1064 nm and 1550 nm. The values of $n_e, n_o$ at $\lambda = 589$ nm for $t=22^\circ$C were obtained from EM Industries, Inc. We fit the obtained experimental points using Cauchy’s dispersion formula:

$$n_{\epsilon,o} = a_{\epsilon,o} + \frac{b_{\epsilon,o}}{\lambda^2} + \frac{c_{\epsilon,o}}{\lambda^4}.$$

(3.11)
Fig. 3.12. Transmission spectra of the pair of $45^0$ TNs with opposite handedness arranged in accordance to Fig.3.10 and Fig.3.11a. Squares: experimental measurements, solid line: calculations; analyzer is oriented parallel to the projection of $\mathbf{n}$ set at the output substrate of TN2 onto the $xy$ plane. Triangles: experimental measurements, dash line: calculations; analyzer is oriented perpendicular to the projection of $\mathbf{n}$ set at the output substrate of TN2 onto the $xy$ plane.

The fitting results returned the following values of the fitting parameters:

$$a_e = 1.67, \quad b_e = 5.46 \cdot 10^{-15} m^2, \quad c_e = 4.24 \cdot 10^{-27} m^4,$$
The normalized optical transmission of the designed polarization rotator is rather high \( T/T_0 > 87\% \) in the wide spectral range \( 0.5 \mu m \leq \lambda \leq 2.7 \mu m \), Fig.3.12. Here \( T \) is the transmission of the pair of \( 45^0 \) TNs sandwiched between polarizers, \( T_0 \) was measured as follows: two \( 20 \mu m \) \( 45^0 \) TN cells were filled with immersion fluid of \( n_D^{25^0C} = 1.6 \) (Cargille\textsuperscript{TM}) and placed between parallel polarizers. The transmission \( T_0 \) of the system was measured in a spectral interval \( 0.5 \mu m \leq \lambda \leq 2.7 \mu m \) and used for the normalization of the transmission of \( 45^0 \) TNs filled with MLC-2048. The optical transmission of the system is similar when the driving signal is reversed: TN1 is under \( U_{20 \text{ kHz}}^{\text{hold}} = 5 \text{ V} \) while TN2 is under \( U_{1 \text{ kHz}}^{\text{hold}} = 50 \text{ V} \) as sketched in Fig.3.11b. The optical signal experiences some additional oscillations (Fig.3.12), which may be caused by Fresnel reflections at the interfaces; the issue can be addressed by using antireflection coatings. Overall, the intensity of the unwanted light polarization is higher than the calculated data; this difference is more pronounced in visible and near infrared regions. This behavior of the transmitted signal is most probably caused by the residual director distortions near the surfaces that transform the incident linear polarization into the elliptical one and, thus, lead to the leakage of undesired polarization of light. This leakage apparently is more pronounced for shorter wavelengths. In \( 45^0 \) TNs, the parasitic effect can be mitigated by further optimization of cell parameters such as the twist angle and adjustments of mutual orientation of the cells.
3.4 Conclusions

We demonstrated applicability of SmA materials in single birefringent prisms and arrays. SmA elements can be used in non-mechanical DBDs that are based on decoupled pairs of electrically-controlled liquid crystalline polarization rotators, such as TN cells and passive deflectors. This approach allows one to separate the issues of response time and beam deflection angles and optimize these two parameters separately.
The deflection angles can be optimized by the design of the birefringent prisms. SmA-filled prisms are attractive in low-cost applications where one needs large apertures, large angles of deflection, and/or non-trivial geometries. We demonstrated that mixtures of homologues of 4,4’-n-dialkylazoxybenzene produce SmA phases with a broad temperature range of SmA existence (up to 30°C for binary mixtures) with a relatively small number of residual defects, such as FCDs, and high transmission characteristics. We determined the typical magnetic fields needed to remove director distortions around the mechanical inclusions and FCDs. Magnetic alignment is most effective when the material is aligned in the nematic phase and then cooled down to the SmA phase.

The SmA-filled birefringent prisms have certain advantages as compared to the crystalline prisms. The SmA prisms are easier and cheaper to form. The optical axis of SmA prisms can be controlled by surface alignment. They can be prepared as relatively thick prisms (up to 2-5 mm in our case) or as arrays of (micro) prisms. Light scattering in SmA birefringent prisms can be reduced by proper alignment to levels that are significantly lower than light scattering at the director fluctuations in the nematic samples of the same thickness. As the light scattering is caused mostly by FCDs that have a fixed size, it becomes smaller with the increase of the wavelength of light; the IR part of the spectrum is less sensitive to these losses. Thus, the SmA prisms are suitable candidates for beam steering not only in the visible part of the spectrum, but in the infrared part as well. An obvious drawback of the SmA prisms is that they can be used only within the temperature range of the SmA phase. The latter can be expanded significantly by using mixtures, as in this work.
To complete the deflection pair of elements usually used in DBDs we developed a broadband polarization rotator based on two 20 $\mu$m thick 45° TNs with opposite handedness. Implementation of a high pretilt angle of $n$ near the boundaries, dual-frequency feature of the LC material and overdriving technique allows one to significantly reduce the switching time of the rotator down to 2 ms. The rotator showed high transmission $>87\%$ in a wide spectral range $0.5 \mu m \leq \lambda \leq 2.7 \mu m$ and can be used in DBDs or in optical elements where fast rotation of the polarization state of multiband light is required.

### 3.5 References


Chapter 4

Design of tunable lens based on dual-frequency nematic

4.1 Introduction

Optical lenses are among the key optical elements. They are widely used in science, industry and our daily life. Development of tunable lenses with a variable focal length is of great importance for a number of applications, ranging from eyeglasses with the adjustable focal length for vision correction to fast non-mechanical zooming devices in photo cameras and camcorders. Lens effect can be achieved when the optical paths of the light beams propagating through the different parts of a medium are different. Thus, the lens can be formed either by varying the thickness of the material while preserving the refractive index of a medium constant through the sample \( n = \text{const} \) (as in the conventional glass lenses, Fig.4.1a), or by varying the refractive index \( n(x, y, z) \) across a flat sample, Fig.4.1b. In both cases for perfect (stigmatic) imaging the quantity \( \int_{P_1}^{P_2} n(x, y, z) dS \) or the integral over all optical paths \( S_1, S_2, ..., S_n \) between two conjugate points \( P_1 \) and \( P_2 \) should be identical.

One of the actively pursued approaches for tunable lens is based on the effect of reorientation of nematic liquid crystal (NLC) in the external electric field. NLCs possess large optical birefringence and dielectric anisotropy; as a result, focusing
The properties of NLC-based lenses can be changed in a wide range with a rather fast response to the applied electric field.

The development of NLC lenses started in late 1970s when a NLC cell of varying thickness with conductive electrodes deposited on the curved substrates was proposed [1, 2]. Subsequent designs used NLC cells of a constant thickness but with patterned electrodes to achieve a non-uniform distribution of the refractive index by electrical addressing of multiple electrodes [3-5], Fresnel-type lenses filled with LCs [6-8], LC lenses with spatially varying density of a polymer network [9-12], lenses formed by patterning of a solid electrode [13-18], and other approaches [19-21].

One of the fundamental problems in the development of electrically-controlled NLC lenses is their relatively slow response. The NLC lens needs to be rather thick to achieve a sufficiently wide focus variability. However, by increasing the thickness $d$ of the lens, one significantly increases the time needed for director reorientation, as the
latter scales as $d^2$ [22]. The typical estimate time of the director reorientation is the quantity $\tau_{\text{off}} = \gamma_1 d^2 / \pi^2 K$ [22], see Eq.3.9. $\tau_{\text{off}}$ describes how fast the director relaxes back to the original state when the field is switched “off”. For example, a cell of thickness $d=100 \, \mu m$ filled with a typical NLC material with $K = 10^{-11} \, N$, $\gamma_1 \approx 0.1 \, kg \, m^{-1} \, s^{-1}$ [22] needs about $\tau_{\text{off}} \approx 10 \, \text{sec}$ to relax the director. The switch-on state can be estimated as $\tau_{\text{on}} = \frac{\gamma_1 d^2}{-\pi^2 K + \epsilon_0 \Delta \epsilon U^2}$, where $U$ is the applied voltage.

Therefore, $\tau_{\text{on}}$ can be reduced by applying the electric field of large amplitude; however, the same principle cannot be applied to reduce $\tau_{\text{off}}$. The problem has been addressed in the past by using thinner cells or polymer-stabilized NLC [9-12]. Thinner cells, however, result in a reduced optical power of the lens, while adding the polymer component might cause a light scattering.

In this Chapter we report on an electrically-controlled NLC-based optical lens filled with a dual-frequency nematic material [23]. The lens design employs a hole-patterned electrode structure in a flat nematic cell. In order to decrease the lens switching time we maximize the dielectric torque by using a dual-frequency nematic material that is aligned at an angle about $45^0$ with respect to the bounding plates by obliquely deposited SiO$_x$ and by using an overdriving scheme of electrical switching [24]. Depending on the frequency of the applied field, the director $n$ realigns either toward the homeotropic state (perpendicular to the substrates) or toward the planar state (parallel to the substrates),
which allows one to control not only the absolute value of the focal length but also its sign. Optical performance of the designed LC lens is close to that of an ideal thin lens.

4.2 Design of electrically tunable lens

We construct the dual-frequency NLC lens from two different glass plates. One plate is 1.1 mm thick with a continuous transparent indium tin oxide (ITO) electrode. The second substrate is 0.2 mm thick with a hole-patterned aluminum electrode (Fig.4.2). To ensure the optimum lens properties, the ratio of the lens diameter $D$ to the lens thickness $d$ should be between 2 and 3 [25]. In our case the lens aperture, i.e. the diameter of the hole $D=300 \, \mu m$ while $d = 110 \, \mu m$. The cell is filled with the dual-frequency NLC material MLC-2048. The dual-frequency properties of MLC-2048 were discussed in Sections 3.3.1 and 3.3.2. To electrically address the designed lens we used two characteristic driving frequencies, $f = 1$ kHz, at which $\Delta \varepsilon = 3.2$ and $f = 50$ kHz, at which $\Delta \varepsilon = -3.1$ (both values of the dielectric anisotropy correspond to the temperature 20°C). By addressing the dual-frequency material at two frequencies we control both $\tau_{\text{off}}$ and $\tau_{\text{on}}$ times, see Eq.(3.10). The optical birefringence of the material $\Delta n = 0.22$ at $\lambda = 589$ nm. The initial orientation of the director is set at about $45^0$ with respect to the bounding plates by treating the substrates with an obliquely deposited layer of SiOx [24, 26, 27] to decrease the response time of the device. Though a high pretilt alignment of the LC director leads to an optical power loss in comparison with the planar (low pretilt) geometry, a high pretilt angle plays the key role in realizing both positive and negative lens in the same cell through the change of the frequency of the applied field.
The hole-patterned electrode provides a nonlinear distribution of the electric field inside the LC layer, which causes a non-uniform reorientation of the LC director and thus the lens effect [15, 25]. The light beams linearly polarized along the $x$ axis and propagating through different regions of the lens would experience different phase shift $n_{ef}d$, where $n_{ef} = \frac{n_o n_e}{(n_o^2 \cos^2 \theta + n_e^2 \sin^2 \theta)^{1/2}}$ is the local field-dependent effective refractive index of the LC, $\theta$ is the local field-dependent angle between the direction of light polarization and LC director, $n_o$ and $n_e$ are the ordinary and extraordinary LC refractive indices, respectively.

Fig. 4.2. Design of the hole-patterned lens filled with dual-frequency nematic.
4.3 Electro-optical characteristics of the designed lens

4.3.1 Response time measurements

The experimental set-up for the response time measurements is shown in Fig.4.3. A light beam from a He-Ne laser (\(\lambda=633\text{nm}\)) passes through the beam expander and polarizer \(P\) oriented parallel to the projection of the LC easy axis onto \(xy\) plane, then hits the LC lens and is focused by the glass lens \(L\) onto the optical detector connected to the oscilloscope (Tektronix TDS 210). If no field applied, the diaphragm with an aperture of 250 \(\mu m\) in front of the detector cuts a significant part of the laser beam and the optical signal is minimum. Under electric field at 1 kHz a converging LC lens is induced and the light beam passes through the diaphragm causing an increase in the light intensity measured by the detector. In Fig.4.4a we show the optical response of the designed lens (top trace) under the applied electrical signal (bottom trace). The applied electric field is a sequence of high amplitude pulses accelerating the initial stage of director reorientation and low amplitude holding pulses at two frequencies (1 kHz and 50 kHz) [24] described also in Section 3.3.3.

The dynamics of the lens response is comprised of fast and slow components. Initially, the lens shows a fast response time of about 50 ms during the transition to the focusing state (Fig.4.4b), but then the optical signal experiences a slow drift over about 400 ms, during which the optical signal changes by about 10-15%. The reason for this drift might be a backflow effect and dielectric heating [28]. The same phenomenon occurs when the lens relaxes back to the non-focusing state (Fig.4.4c): a fast reorientation during 75 ms is followed by a slow drift of about 400 ms.
4.3.2 Lens optical performance

In this Section we discuss the optical characteristics of the designed LC lens and compare its performance with an ideal thin lens.

In a cell with a non-zero surface pretilt, the angle between the electric fringe field and LC director is different [29] at the antipodal points of the hole-patterned electrode, Fig.4.5. As the result, the pattern of the field-induced director reorientation and effective refractive index across the lens is nonsymmetrical; the interference pattern shifts with respect to the lens axis. In order to correct the off-axis focusing, we propose a double
Fig. 4.4. The response time of the 110 µm thick lens. (a): Lens optical response (top trace) under applied voltage (bottom trace). The driving signal (bottom trace) is a sequence of high amplitude pulses (50V RMS, 1 kHz) followed by a holding voltage (4V RMS, 1 kHz) followed by high amplitude pulses of 40V RMS at 50 kHz. (b): Transition to the focusing state (top trace). Electric signal applied (bottom trace) is a sequence of high amplitude pulses (50V RMS) followed by a holding voltage (4V RMS) at 1 kHz; (c): Relaxation from the focusing state to the uniform (non-focusing) director state (top trace). The transition is triggered by high amplitude pulses of 40V RMS at 50 kHz (bottom trace).

lens design shown in Fig.4.5. Two identical hole-patterned lenses are coaxial and assembled in a “head-to-head” fashion. The symmetry of this configuration is such that the difference between the direction of the fringe field and the easy axes of two lenses are
Fig. 4.5. The double lens design. Two hole-patterned NLC lenses are coaxial and oriented in a “head-to-head” fashion. The simplified ray paths and director distribution are shown for the field applied at 1 kHz. The lines of electric fringe field are shown by thin lines. The off-axis focusing of the light caused by the first lens is corrected by the second lens.

mutually compensated. In Fig.4.5 we show schematically that by adjusting the voltages $U_1$ and $U_2$ applied to the lenses 1 and 2 we can correct the off-axis focusing of the single lens. We confirmed this in the following experiment.

We placed the double LC lens between crossed polarizers in such a way that the projection of the LC easy axis on the $xy$ plane is at $45^0$ to axes of both polarizers. By using a movable CCD camera with an objective we can monitor the output aperture plane of the lens as well as its focal plane. The experimental set-up is similar to that shown in
Fig.4.3 except now we placed the lens between two crossed polarizers. Due to the dual-frequency properties of the nematic material and a high pretilt alignment we can realize both positive and negative lens regimes by applying the electrical field with frequencies 1 kHz and 50 kHz, respectively.

Under electric field we observe the appearance of the fringes (circles) caused by the interference of the ordinary and extraordinary light waves experiencing a phase shift while propagating through the LC. The number of circles indicates the optical power of the lens. With voltage increase the number of circles and the lens optical power increase too. If \( U_1 = 0 \) the observed interference fringes are distorted and off-centered (Fig.4.6, top row), so the lens focal point is shifted away from the lens axis. After adjusting \( U_1 \), the fringes are becoming well-centered (Fig.4.6, bottom row), so the focal point shift is corrected. The results of correction are shown for both frequencies applied. The focal point shift can be used as a desirable feature in beam deflecting devices. Note, that the optical power of the double lens is larger as compared to the single lens. When \( U_1 \neq 0 \), the number of fringes and lens optical power increase (Fig.4.6, bottom row).

As the thickness of the designed NLC lens is 110 \( \mu m \) and the focal length is of the order of millimeters, we can neglect the displacement of the light beam inside the LC layer. In this case the NLC lens can be considered as a thin lens introducing a phase shift to the incident wave. The optical field of a plane wave \( A_L \) behind the thin lens is described by:

\[
A_L(x, y) = \exp[i k d] \exp[-i \frac{k}{2 f_L} (x^2 + y^2)],
\]  

(4.1)
where  \( k = \frac{2\pi}{\lambda} \) is the wave number (\( \lambda \) is the wavelength), \( n \), \( d \), and \( f_L \) are the lens refractive index, thickness and focal length, respectively [30]. The first exponent describes a constant phase shift \( \phi_0 \), introduced by a thin lens while the second exponent is a quadratic approximation of a spherical wave. Thus, the phase retardation \( \phi \) of the
NLC lens along the $x$ axis (direction of the initial easy axis projection on the plane of the cell) can be written as follows:

$$\phi = \phi_0 - \frac{2\pi}{\lambda} \frac{1}{2f_L} x^2. \quad (4.2)$$

The phase retardation of LC lens at different voltages can be found from the number of circles appeared. The distance between two neighboring dark (or white) fringes corresponds to the phase retardation of $2\pi$. In Fig. 4.7 we show the experimental results of phase retardation measurements (dots) of the double lens together with the fitting curves (the solid lines) at two frequencies of the applied electric field. The fitting curves were obtained from Eq. (4.2) with the focal length $f_L$ as a fitting parameter. The fitting curves are very close to the experimental data; it means that for each voltage applied, the induced NLC lens behaves closely to the thin lens with a certain focal length. Some deviation of the experimental data from the curves describing the behavior of an ideal thin lens indicates the presence of aberrations in the system, such as coma and astigmatism [31]. The aberrations are most probably caused by a nonsymmetrical distribution of the refractive index with respect to the geometric axis of the system. In Fig.4.8 we show the focal length dependence of the double NLC lens versus the applied voltage. Focal length values were extracted from the results of the quadratic fitting. We verified them independently by moving the CCD camera (Panasonic GP-KR222) along the $z$ axis and by imaging a real object (microscope micrometer), Fig.4.9. We placed the microscope micrometer behind the double lens and observed the image formation with electric signal applied at high and low frequencies under the microscope (Nikon
Fig. 4.7. Phase retardation measurements (dots) of the double NLC lens and the quadratic fitting curves (the solid lines) at two frequencies of the applied electric field. (a): 50 kHz; (b): 1 kHz.

Optiphot 2-Pol). Under $U_1=2.9V$, $U_2=3.3V$, $f=50$ kHz the double lens forms an erect reduced virtual image, Fig.4.9b. Under $U_1=1.5V$, $U_2=2.2V$, $f=1$ kHz the double lens forms an erect enlarged virtual image, Fig.4.9c. The schematic image formation is shown in the top row of Fig.4.9. Note, that in both cases the object is located between the lens center and focal point.

The obtained results confirm that both negative and positive lens for the very same nematic cell can be realized by choosing the frequency of applied voltage at either
Fig. 4.8. Focal length of the double LC lens versus the applied voltage at two frequencies. (a): 50 kHz; (b): 1 kHz.

50 kHz or 1 kHz (Figs. 4.7-4.9). The focal length varies from $-\infty$ to -1.9 mm (the optical power varies from 0 to -520 diopters) at 50 kHz and from $+\infty$ to +2.5 mm (the optical power varies from 0 to +400 diopters) at 1 kHz. As the applied voltage increases, the lens focal length decreases (Fig.4.8). At some critical voltage the electric fringing field effect causes the director reorientation in the central part of the lens due to elastic properties of LC. As a result, the optical retardation and focal length of the lens both saturate. With further voltage increase the NLC lens becomes distorted.
Fig. 4.9. Image formation by the double lens DL. Object O (objective micrometer) is between the lens center and focal point f. (a): No voltage applied. (b): High frequency ($f = 50$ kHz) voltage is applied. The induced negative lens forms an erect reduced virtual image i. (c): Low frequency ($f = 1$ kHz) voltage is applied. The induced positive lens forms an erect enlarged virtual image. Top row: the schemes of image formation for both cases are shown.

4.4 Discussion

Let us discuss the advantages of the proposed NLC lens design. First, a combination of the overdriving scheme with a high pretilt angle $\approx 45^\circ$ results in strong restoring torques that facilitate director reorientation from the $45^\circ$ tilted state toward both homeotropic and planar states [24], which in turn reduces the response time during focus
change down to at least 400 ms for the 110 \( \mu m \) thick cell. It is more than one order of magnitude faster than in regular cell designs with \( \tau_{\text{off}} = 10 \text{ sec} \).

Second, a tilt of the LC molecules eliminates the threshold of reorientation allowing a continuous tuning of the optical power of the double LC lens from -520 to +400 diopters in the same cell as illustrated in the example above with \( \lambda = 633 \text{ nm} \).

Third, a high pretilt alignment of the LC molecules allows us to avoid the appearance of disclination lines that may cause light scattering and slow relaxation during lens switching. To eliminate the disclination line appearance in a planar cell with low pretilt angle one can apply an in-plane electric field to force the molecules rotate in the same direction [29]. In our approach no additional field is required; the problem of disclination lines appearance is solved by the high pretilt alignment of LC molecules.

By varying the voltage applied to both cells in the double lens approach, we can focus the light not only along the lens axis, but also move the focus along a 3D trajectory or in the \( xy \) plane, which was realized earlier for planar cells through the application of the in-plane field [17]. This feature may be used in beam deflecting devices.

Note, that the actual response time of our device is determined by a fast component of the order of 50-100 ms with a subsequent slow drift about 400 ms for 110 \( \mu m \) cell. The slow relaxation (Fig.4.4a) is most probably caused by material flow and dielectric heating. Slow drift of the optical signal can be reduced by either polymer stabilization or using the thinner cells (of course in the latter case the LC lens will be less powerful).
The proposed lens design is polarization-dependent, which means that only the light with polarization parallel to the easy axis will be focused while the light with orthogonal polarization will remain unfocused. If necessary, the polarization dependency can be eliminated by stacking LC cells with the easy axes oriented perpendicularly to each other [32]. Note, that in our experiments, we used cells with a relatively small aperture of 0.3 mm. For applications in photocameras and camcorders the LC lenses should be few times larger. The techniques to upscale the LC lenses to the 1-5 millimeter-range aperture have been described, for example in Ref. [16]. These techniques are applicable to the dual-frequency design proposed in this work.

4.5 Conclusions

We have demonstrated a dual-frequency NLC-based lens with a hole-patterned electrode structure, which focal length can be tuned by the electric field. The proposed design allows one to use the same cell as either a negative or positive lens simply by changing the frequency of the applied field. Note, that the realization of positive and negative lens in one design was shown earlier by utilizing, for example, a combination of dual-frequency nematic aligned tangentionally and plastic Fresnel lens [8] or by applying two driving voltages in a hole-patterned design [33]. Our approach combines the high pretilt alignment, dual-frequency properties of the nematic material and overdriving scheme and allows us to reduce the response time during focus change from about 10 sec (conventional scheme) down to 0.4 sec for a 110 \( \mu m \) thick cell; most of the director structure is switched within the first 50-100 ms. The optical characteristics of the system
are close to the performance of an ideal thin lens but some wavefront aberrations are still present. The optical power of the system with the aperture $D=300\mu m$ can be continuously tuned from -520 to +400 diopters ($\lambda = 633\text{ nm}$) in a double NLC lens approach. Such a design can be used to achieve fast optical communication between multiple channels, for example in microlens arrays [34-37], in beam steering or scanning devices or for fast non-mechanical zooming in miniature cameras. In the latter case isotropic fluids have been recently proposed as the focusing medium [38, 39]. Isotropic liquid lenses are based on electrowetting effect, which allows one to control the droplet or the meniscus shape by applying the electric field. If the densities of two liquids in contact are equal, the meniscus is rather insensitive to the external vibrations and shocks [39]. The advantage of the NLC lenses, however, is in a greater mechanical stability against vibrations as the nematic fluid is confined in a micrometer-thick gap between two rigid plates; the refractive changes are caused by molecular reorientation and not by the variation of the fluid surface/interface.

4.6 References


CHAPTER 5

Electric field-induced dynamics of colloidal particles controlled by a backflow

5.1 Introduction

Behavior of particles dispersed in a host medium either in a form of emulsion where foreign liquid droplets stabilized by surfactant are scattered in a liquid medium or in a form of colloidal dispersions where the foreign particles are solid, or aerosols with liquid or solid particles scattered in a gaseous medium, are of significant research interest. The interest is caused by usage of colloidal dispersions in food industry, personal care products, paints, pigments and inks. Besides consumer applications, colloidal dispersions are excellent model systems to study viscoelastic properties of complex fluids, Brownian motion, interparticle interactions, aggregation, hydrodynamics, etc. Colloidal systems show rather rich and interesting phenomena. For example, attractive interactions may lead to clustering of the colloidal particles, thus changing the properties of the system and product shelf life [1]. An applied electric field can cause rotational and translational motion of small particles dispersed in a fluid [2] allowing also for particle manipulation [3]; application of a modulated field may lead to appearance of jammed structures and ordered networks of colloidal particles [4]. Recently, the designs of thin electronic paper displays were announced utilizing absorbing pigments and based on electrophoretic [5, 6] and electrowetting effects [7].
In LC-based colloidal dispersions the observed phenomena are further enriched. In addition to the well-known interparticle forces, such as electrostatic and van der Waals forces, and the entropy (excluded volume) effects, LC colloids demonstrate fascinating elasticity-mediated interactions [8, 9]. The introduction of colloidal particles into uniform nematic may lead to the appearance of director distortions in the bulk due to anchoring of LC molecules at the particle’s surface. The character of distortions depends on particle size and anchoring strength and can be of dipolar or quadrupolar type depending on symmetry of the director distribution around the particle [9, 10]. Director distortions induce additional long range particles interactions absent in isotropic fluids [11, 12]. The underlying physics is based on the unusual features of the elastic free energy and surface energy density of the LC. A particle of a typical radius $R$ embedded in the nematic matrix is associated with the energy of director distortions that scales as $KR$ and the surface anchoring energy that scales as $2WR^2$. Here $K$ is the Frank elastic constant of director distortions and $W$ is the (polar) anchoring energy. Clearly, if the particle is larger than some critical radius $R_c \sim K/W$, it will create director distortions in the surrounding LC. When the preferred orientation of the director is along the normal to the particle/LC interface, then the particle creates a defect of a dipolar type, Fig.5.1. A small particle, $R \ll K/W$, would not distort the director field much.

The case of $R \gg K/W$ represents the most dramatic departure of the LC colloids from their isotropic counterparts, as the elastic interactions bring a decisive long-range contribution to the interparticle interactions. These interactions cause rather fascinating phenomena such as ordering of colloidal particles in chains [8, 9] and hexagonal...
Fig. 5.1. Schematic of colloidal particle in nematic LC. (a): Small particle does not disturb LC director. (b): Particle with the size larger than some critical radius $R_c \sim K/W$ and normal boundary conditions creates a satellite defect, a hyperbolic hedgehog. Note the dipolar symmetry of the overall director distortions around the particle.

Since LCs are anisotropic dielectrics and anisotropic (weak) conductors, it is natural to expect that the LC colloids should also exhibit a number of interesting field-induced effects. Recent studies demonstrated that the electric field can cause translational and orientational motion of colloids in the LC medium [16-21]. Reorientation of LC molecules by the external field leads to backflow, which was nicely visualized by Zou and Clark for the case of ferroelectric SmC [22]. Earlier in Chapters 3 and 4 we showed...
that the electric field-induced backflow slows the response time of LC devices causing a
transient dynamics of the output optical signal. However, as numerous studies reveal that
direction and velocity of backflow depend on cell geometry and director configuration
[23-30], a backflow effect opens the possibility for electrically-driven particles
manipulation in anisotropic medium [16, 17, 21]. This may result in many practical
applications, including particles sorting, electrically-driven LC microfluidic devices,
development of LC-based electrophoretic displays, etc.

The central question in the rapidly growing field of LC colloids is the role of the
director distortions, especially in the dynamic phenomena. In this Chapter we present our
studies of the dynamics of colloidal particles caused by the electric field in a
homogeneously aligned LC cell [16, 17]. First, we quantify the elastic repulsion forces
between a heavy colloidal particle and the bounding walls of the LC. Interestingly, the
elastic “buoyancy” force that keeps the particle in the nematic bulk, grows with the
particle size as $R^4$; i.e., faster than the gravity force $R^3$. As a result, a larger particle
finds itself farther away from the bottom of the cell as compared to a smaller particle.
Second, we discuss the lifting forces that move the particles towards the bounding plates
in the presence of the electric field. Here, two mechanisms are possible: an elastic
entrapment similar to the one described in [14] and dielectrophoresis (or Kelvin forces)
discussed in [31-33]. Finally, we analyze the backflow-induced bidirectional motion of
the particles perpendicular to the applied field direction, which is in contrast to the
parallel motion observed for electrophoresis. In our studies we use the fluorescence
confocal polarization microscopy (FCPM) technique [34] to determine director
orientation around the particles and to trace the particles location and movement. The velocity of particles agrees well with the idea that their dynamics is controlled by the backflow effect.

5.2 Interactions of particles with bounding walls: effect of levitation

We studied the mixtures of nematic LC E7 (EM Industries, Inc.) with small concentrations of colloidal particles (<1% by weight). The surface of particles was modified by octadecyltrichlorosilane (Sigma-Aldrich) to promote normal boundary conditions of director \( \mathbf{n} \) at particle’s surface. To modify the surface we dispersed some quantity of colloidal spheres in the same amount of mixture hexane/octadecyltrichlorosilane (1% wt.). After heating and evaporating of hexane, 4.9 \( \mu m \) and 9.6 \( \mu m \) in diameter glass particles (Duke Scientific) were dispersed in E7 doped with a small amount (0.01 wt. %) of fluorescent dye N,N'-Bis(2,5-di-tert-butylphenyl)-3,4,9,10-perylenedicarboximide (BTBP, Sigma-Aldrich). We filled the cells formed by glass substrates covered with ITO and rubbed polyimide PI2555 layers with the obtained mixture to get a homogeneously aligned nematic slab; rubbing resulted in a small (1-2°) pretilt angle of the director.

Both polarization microscopy (PM) and FCPM studies revealed a formation of hyperbolic hedgehogs accompanying each colloidal particle (Fig.5.2) and thus confirming normal director orientation at particle’s surface. When no electric field is applied the colloidal particles are levitating in the bulk. Interestingly, the heavier 9.6 \( \mu m \)
particles are located closer to the middle of the cell than the smaller 4.9 \mu m \text{ particles (Fig.5.2k,l). Let us discuss this in more detail.}

At the absence of field the colloidal particles accompanied by defects of a dipolar type are floating in the bulk balanced by the gravitational force $F_g = \frac{4}{3} \pi R^3 (\rho_p - \rho) g$ and the repulsive forces of dipole-dipole interactions, which potential can be written as [8]:

$$E = 4\pi K p_x \frac{1 - 3 \cos^2 \phi}{r^3}. \quad (5.1)$$

Here $K = 15 \text{ pN}$ is the average of splay $K_1 = 11.7 \text{ pN}$ and bend $K_3 = 19.5 \text{ pN}$ elastic constants of E7 [35]; $p_x$ is the component of the dipole moment $p$ of a hedgehog along the $x$ axis, Fig.5.2. In our case $p \parallel x$, then $p_x = AR^2$, $A = 2.04$ is the numerical constant [8]; $R$ is the particle’s radius; $\phi$ is the angle between the vector $\mathbf{r} = \mathbf{r}_1 - \mathbf{r}_2$ and the $x$ axis; vectors $\mathbf{r}_1$ and $\mathbf{r}_2$ are describing positions of two defects (for our geometry $\cos \phi = 0$ as $\mathbf{r} \perp x$); $r$ is the distance between two dipoles, $r = |\mathbf{r}|$; $\rho_p \approx 2.5 \text{ g/cm}^3$ (Duke Scientific) and $\rho \approx 0.98 \text{ g/cm}^3$ (EM Industries, Inc.) are the densities of the glass particle and E7, respectively; $g \approx 9.8 \text{ m/s}^2$ is the standard gravity. The balance of forces acting on the particle accompanied by hyperbolic hedgehog can be found by the method of images (Fig.5.3a) as follows:

$$F = F_0 - F_h - F_g = \frac{3}{2} \pi K A^2 \left( \frac{2R}{h - 2\delta} \right)^4 - \frac{3}{2} \pi K A^2 \left( \frac{2R}{h + 2\delta} \right)^4 - \frac{4}{3} \pi R^3 (\rho_p - \rho) g = 0. \quad (5.2)$$
Fig. 5.2. FCPM studies of the director structure around glass spherical particles (4.9 \( \mu m \) and 9.6 \( \mu m \) in diameter) with normal boundary conditions in a \( h = 33 \mu m \) thick planar cell. (a, b): Schemes of director distribution around the particles; hyperbolic hedgehogs with different orientation are observed: left-oriented hedgehogs (left column) and right-oriented hedgehogs (right column). Orientation of the polarizer \( P \), director \( n \) and coordinate axes are shown. (c-f): Transmission mode, crossed polarizers. (g-j): Fluorescent mode, scan in the \( xy \) plane. (k-n): Fluorescent mode, scan in the \( z \) direction. No field is applied; particles are levitating in the bulk. Distances from the big (\( \delta_1 \)) and small (\( \delta_2 \)) particles to the middle of the cells are shown. Bigger particle is found closer to the middle than the small one (\( \Delta \delta = \delta_2 - \delta_1 \sim 2 \mu m \)).
Here $F_0 = \frac{3}{2} \pi K A^2 \left( \frac{2R}{h - 2\delta} \right)^4$ is the repulsion force between the particle and its mirror image created by the bottom $z = 0$ plate; $F_h = \frac{3}{2} \pi K A^2 \left( \frac{2R}{h + 2\delta} \right)^4$ is the repulsion force between the particle and its mirror image created by the upper $z = h$ plate (Fig. 5.3a); $h$ is the cell thickness, $\delta$ is the shift of the particle’s center from the middle $z = h/2$ of the cell. Forces $F_0$ and $F_h$ can be found by differentiating the potential $E$ with respect to $r$, see Eq.(5.1). We neglect the effect of higher order mirror images.

The particle is floating in the bulk repelled by the boundaries, Fig.5.3b,c. We verified the position of particles in the experiment by scanning the cells of different thickness in the $z$ direction using the FCPM technique, Fig.5.4a. The experimental error of measurements of $\delta$ related to the aberrations of light propagating through the birefringent medium in FCPM scans can be mitigated through the introducing an appropriate correction factor, if necessary, by knowing a) the thickness of the cell; b) the scan distance along the $z$ axis; c) the size of particles. Calculated values of $\delta$ can be found by solving the Eq. (5.2) with respect to $h$. Both calculated and experimental dependencies of $\delta$ versus $h$ for particles of diameters $4.9 \mu m$ and $9.6 \mu m$ are shown in Fig.5.4b. $\delta < 1 \mu m$ if $h < 20 \mu m$ for both particles and monotonously grows with thickness increase. Note, that larger (and heavier) particles are closer to the middle of the cell than the smaller ones, which is rather counterintuitive.
Fig. 5.3. Schematic of the forces acting on the particle accompanied by the hyperbolic hedgehog. (a): The particle is floating in the bulk balanced by the repulsive forces from the boundaries. (b): FCPM image of $2R = 4.9\mu m$ glass particle floating in LC bulk and accompanied by a satellite defect. (c): Sketch of director distribution and forces (gravitational $F_g$ and repulsion ones $F_0, F_h$) acting on a particle.
Fig. 5.4. (a): FCPM $z$ scans of LC cells. Glass particles of 4.9 $\mu$m and 9.6 $\mu$m in diameter are floating in the bulk. The thicker the cell the closer the particles are to the bottom substrate. Note, that bigger particles are closer to the middle of the cell than the smaller ones. (b): Plot demonstrating experimental and calculated dependency of the shift $\delta$ versus cell thickness. $\delta$ is calculated according to Eq.(5.2) and describes the displacement of the glass particles of 4.9 $\mu$m and 9.6 $\mu$m in diameter dispersed in E7 from the middle of the cell.
5.3 Effect of lifting forces: selective movement of particles

As we clarified in the previous Section, at the absence of field the particles accompanied by hedgehogs are floating in the LC bulk, Fig.5.5a-f. Under electric field \( \mathbf{E} \parallel z \) when the voltage exceeds the Frederiks threshold \( U_{th} = \pi \frac{K_1}{\varepsilon_0 \Delta \varepsilon} \approx 0.92 \text{ V} \) [26], the LC director reorients and the particles move toward the substrates, Fig.5.5g-j. Here \( \varepsilon_0 \approx 8.85 \times 10^{-12} \text{ C}^2 / \text{N} \cdot \text{m}^2 \) is the permittivity of free space, \( \Delta \varepsilon = \varepsilon_{||} - \varepsilon_{\perp} = 19.0 - 5.2 = 13.8 \) (EM Industries, Inc) is the dielectric anisotropy of E7. We applied the electric signal with the rate of the voltage envelope increase set low, 0.1 V/s to minimize the backflow. The carrier frequency of the signal was set high, \( f_c = 10 \text{ kHz} \), to avoid electrohydrodynamics caused by ions. Both pretilt angle and the orientation of hedgehog determine the direction of the upward or downward shift of the particles (Fig.5.5g-j), thus the particles selectively move towards the opposite substrates.

There are two possible mechanisms that may cause a shift of the colloids from the middle of the cell towards the top and bottom plates. The first one is the elastic mechanism [14], associated with the elastic interactions of the director around the colloid and near the surfaces. The second mechanism is of dielectrophoretic origin [31-33]: as the electric field and dielectric permittivity in the distorted cell change along the vertical \( z \) axis, the dielectric sphere (the particle) in a dielectric medium (LC) might experience a force resolved along the \( z \) axis. Below we estimate the typical forces associated with each mechanism.
Fig. 5.5. FCPM studies of the glass particles behavior (4.9 μm and 9.6 μm in diameter, cell thickness $h \approx 33 \mu m$) under applied voltage. (a, b): Schemes of director distribution around the particles accompanied by hyperbolic hedgehogs; (c-f): FCPM scan in the $z$-direction. No field is applied; particles are levitating in the bulk; (g-j): FCPM scan in the $z$-direction. Electric field ($U = 1.3$ V, $f_c = 10$ kHz) is applied. Particles with opposite orientations of hyperbolic hedgehogs are pushed towards the opposite substrates.
Director distortions near the substrates caused by the surface anchoring of \( n \) (the electric field aligns \( n \) perpendicular to the substrates in the bulk while the surface anchoring forces keep \( n \) parallel to the substrates near the boundaries) concentrate over the length \( \xi = \frac{h}{U} \sqrt{\frac{K}{\varepsilon_0 \Delta \varepsilon}} \) \([26]\), where \( U \) is the applied voltage. Elastic energy gained by moving the spherical particle from the uniform middle of the cell towards the distorted regions near the substrates can be estimated as \( \Delta E \sim KR^3 / \xi^2 \) \([14]\); The corresponding trapping force \( F_{\text{trap}} \sim KR^3 / \xi^3 \sim 1.2 - 9.4 \text{ pN} \) is rather significant, as compared to the gravity forces and elastic repulsive forces; e.g., Eq.(5.2) yields \( F_h \approx 0.05 - 1.2 \text{ pN} \), \( F_0 \approx 0.7 - 6.2 \text{ pN} \) for \( h = 33 \mu \text{m}, 2R = 5 - 10 \mu \text{m} \) and \( E \sim 0.06 \text{ V/ \mu m} \).

The dielectrophoretic effect is associated with the different director orientation near the substrates and the middle of the cell. The electric field is a function of the \( z \) coordinate; the dependence can be found from the condition of a constant value of the electric displacement as follows:

\[
E(z) = \frac{E_0 \varepsilon_{\text{eff}}}{\varepsilon_\perp \sin^2 \theta + \varepsilon_\parallel \cos^2 \theta},
\]

(5.3)

where \( \theta \) is the angle between the applied field \( E_0 = e_0 U / h \) and the local director, and \( \varepsilon_{\text{eff}} = h / \int_0^h (\varepsilon_\perp \sin^2 \theta + \varepsilon_\parallel \cos^2 \theta)^{-1} \, dz \). For simplicity, let us assume that the director in the middle of the cell is completely reoriented along the field, so that \( \theta = 0 \), while near the substrates it remains parallel to the substrates, \( \theta = 90^0 \). In this case the values of the electric field in the center of cell and near the substrates (say, the top substrate) are
\(E^{\text{center}} = E_0 \frac{\epsilon_{\text{eff}}}{\epsilon_{\|}}\) and \(E^{\text{top}} = E_0 \frac{\epsilon_{\text{eff}}}{\epsilon_{\perp}}\), respectively. A dielectric sphere with permittivity different from that of the surrounding medium experiences a force in a non-uniform electric field. To roughly estimate this force, we first recall that the polarization of a dielectric sphere of radius \(R\) placed in a uniform electric field \(E_0\) is [31]:

\[
P = 4\pi R^3 \epsilon_0 \epsilon_{\text{LC}} \frac{\epsilon_p - \epsilon_{\text{LC}}}{\epsilon_p + 2\epsilon_{\text{LC}}} E_0, \quad (5.4)
\]

where \(\epsilon_p\) and \(\epsilon_{\text{LC}}\) are the dielectric constants of particle and LC, respectively. The energy of the particle in the uniform field is \(W = -\frac{1}{2} \mathbf{P} \cdot \mathbf{E}_0\). When the particle is transferred from the middle of the cell towards the substrate, the associated energy difference can be estimated roughly as follows:

\[
\delta W = W^{\text{center}} - W^{\text{top}} = -2\pi R^3 \epsilon_0 \epsilon_{\text{eff}}^2 \left[ \frac{\epsilon_p - \epsilon_{\|}}{\epsilon_p + 2\epsilon_{\|}} \right]^2 - \left( \frac{\epsilon_p - \epsilon_{\perp}}{\epsilon_p + 2\epsilon_{\perp}} \right)^2. \quad (5.5)
\]

The corresponding dielectrophoretic force can be estimated as \(F_{\text{diele}} \sim -\delta W / \xi\). With \(\epsilon_p = 5.8\) (Duke Scientific), \(h = 33 \mu m\) \(U = 1.3 V\) and \(\epsilon_{\text{eff}} \approx 8\) (low field regime) we find that the net force is \(F_{\text{diele}} \sim 0.2 \text{ pN}\) for \(2R = 5 \mu m\) and \(F_{\text{diele}} \sim 1.9 \text{ pN}\) for \(2R = 10 \mu m\).

The force is directed from the middle of the cell towards the substrate. It appears that in the low-field regime \(E < 0.06 V / \mu m\), the dielectrophoretic force is weaker than the elastic trapping force near the substrates, so the prime mechanism for particle’s lifting in this case is the elastic one; however, for the higher fields the dielectrophoretic force becomes quickly dominating: for \(E \sim 0.5 V / \mu m\) one finds that \(F_{\text{diele}} \sim 2500 \text{ pN}\).
\( F_{\text{trap}} \approx 700 \text{ pN} \) for \( 2R = 5 \mu m \) and \( F_{\text{die}} \approx 20000 \text{ pN} \) (\( F_{\text{trap}} \approx 5500 \text{ pN} \)) for \( 2R = 10 \mu m \) particles. Both the elastic and dielectrophoretic effects deserve further studies.

Because of the specific antisymmetric character of the backflow in the LC cell with “antiparallel” boundary conditions at the opposite plates [26], particles’ shift towards the top and bottom substrates allows one to establish a bidirectional flow that would carry particles with different orientation of the elastic dipole along two antiparallel directions [16, 17]. Below we discuss the phenomenon in more detail.

### 5.4 Dynamics of colloids observed under modulated electric signal

In this Section we discuss the dynamics of colloidal particles under modulated electric signal (Fig. 5.6) observed in a regular homogeneously aligned LC cell with a small pretilt angle (1-2\(^0\)) of the director.

It is known that director reorientation leads to backflow [22-30]; backflow velocity profile varies for different geometry of LC cell being antisymmetric in planar cells [26], unidirectional in Pi-cells [27, 28], nonsymmetric in hybrid cells [29], 2D in TN cells [21, 30]. Although the dynamic properties of nematic LCs with consideration of backflow were illustrated in many works for different director configuration it is worth to notice that all these investigations are restricted to the simple switching scheme: switching field “on” and/or switching “off”. However, the additional consequences of flow that may result from the complicated switching schemes, such as modulated signals, have received little attention and are poorly understood. In fact, a lot of LC devices work
under modulated signals and require fast “on” and “off” switching. Understanding of the flow profiles and dynamic properties observed under different switching schemes may lead to 1) improvement the device switching characteristics; 2) design microfluidic devices; 3) manipulation of colloidal particles as was demonstrated in TNs [21] and planar cells [16, 17]. We discuss our findings on colloidal dynamics below.

We studied the mixtures of E7 with $2R \approx 4.09\mu m$ silica particles (0.5 wt. %, Bangs Laboratories, Inc.) filled into $h = 21\mu m$ thick planar cells. In LC particles were accompanied by hyperbolic hedgehogs of opposite signs and located approximately in the middle of cell (see Fig.5.4) at the absence of field. The backflow was induced by applying a signal with amplitude $U = 10\,\text{V}$ and carrier frequency $f_c = 10\,\text{kHz}$ (generator DS345, Stanford Research Systems, Inc.) modulated with a frequency $f_m = (0.5-100)\,\text{Hz}$, Fig.5.6. The duty ratio defined as the duration of the field “on” to
the total duration of the field cycle was set at 50%. The modulated signal was turned “on” after the lift-creating pulse described in Section 5.3, which allowed for the spatial separation of the particles according to the orientation of their elastic dipoles towards the top and bottom substrates. The spatial separation offers an opportunity to move these particles along two opposite directions in the plane of the cell by antisymmetric backflow induced by modulated signal that was observed in the experiment, Fig.5.7.

To verify the experimental findings we performed the numerical simulations of the induced backflow. The results are reported in [16, 17]. The modeling of backflow was based on Ericksen-Leslie equations [36-39] of nematodynamics. These equations couple the director reorientation with the hydrodynamic flow of LC fluid. To simplify calculations couple assumptions were made: 1) we assumed that director \( \mathbf{n} = \{\cos \theta(z,t), 0, \sin \theta(z,t)\} \) is in the \( xz \) plane; 2) the hydrodynamic flow has only an \( x \) component, \( \mathbf{v} = \{v(z,t), 0, 0\} \); 3) we neglected the flow inertia term, because the flow relaxation time \( \tau_v = \frac{\rho h^2}{\pi^2 \alpha_4} \sim 1 \mu s \) [26] is much shorter than the director relaxation times \( \tau_{on} = \frac{\gamma_1 h^2}{(\varepsilon_0 \Delta \varepsilon U^2 - \pi^2 K_1)} \sim 10 \) ms and \( \tau_{off} = \frac{\gamma_1 h^2}{\pi^2 K_1} \sim 1 \) s. Here \( \gamma_1 = \alpha_3 - \alpha_2 \) is the rotational viscosity, \( \alpha_2 = -282 \), \( \alpha_3 = -1 \), and \( \alpha_4 = 225 \) (all in \( mPa \cdot s \) units) [35] are the viscosity coefficients, \( \rho \) is the density of E7.
Fig. 5.7. Electric field-induced bidirectional motion of particles 1 and 2. (a): Initial state, $U = 0$, particles at $z \approx h/2$; (b-e): Bidirectional motion driven by $U = 10$ V, $f_c = 10$ kHz, $f_m = 10$ Hz. Particle 1 migrates to the left ($-x$), while particle 2 moves to the right ($+x$). The particles of type 1 and 2 are well separated along the $z$ axis and do not trap each other; $t$ is the time since modulated signal is turned “on”.
The flow behavior is different for $f_m \ll \tau^{-1} = \left( \tau_{on} + \tau_{off} \right)^{-1} \approx \tau_{off}^{-1} \approx 1 \text{ Hz}$, when $n$ has time to equilibrate, and for $f_m \geq \tau^{-1}$, when $n$ does not equilibrate. The regime $f_m \ll \tau^{-1}$ is well studied [26]. When the field is switched “on”, a counterclockwise rotation of $n$ causes a flow with $v_{on}(h/2 < z < h) < 0$ and $v_{on}(0 < z < h/2) > 0$, as shown schematically in Fig.5.8b. With time, this flow gradually vanishes. When the field is switched “off”, elastic torque causes a clockwise rotation of director near the substrates and initially $v_{on}(h/2 < z < h) < 0$, $v_{on}(0 < z < h/2) > 0$, Fig.5.8c. With time the director relaxation in the bulk is causing a slow reverse flow so $v_{on}(h/2 < z < h) > 0$, $v_{on}(0 < z < h/2) < 0$ now, Fig.5.8d.

When $f_m \geq \tau^{-1}$, equilibration after each “on” and “off” switch is not complete. Time average of the backflow for different modulation frequency can be calculated as follows:

$$< v(z) > = \frac{\int_{v_{on}} v(z,t) \, dt}{1/f_m}.$$  \hspace{1cm} (5.6)

The results of calculation are in Fig.5.9. The symmetry of the planar cell dictates that the backflow velocity is zero in the middle of the cell but reaches its maximum near the top and bottom plates; the directions of the mass flow near the top and the bottom plates are opposite to each other. The particles are thus carried bidirectionally, Fig.5.7. The motion is similar to the one observed by Ref. [20], with that difference that in our case it is
Fig. 5.8. Dynamics of director reorientation and backflow. (a): Scheme of the applied modulated signal. (b-d): Sketch of director reorientation when the field is turned “on” and “off”.

Bidirectional rather than a unidirectional. This antisymmetric backflow controlling the bidirectional translation of the particles can be in turn controlled by changing the frequency and modulation profile of the applied field [16, 17].

Let us compare the experimental measurements of particles velocity with calculations of backflow. In the experiment the dependence $\langle v_p \rangle (f_m)$ is non-monotinous, with $\langle v_p \rangle \rightarrow 0$ for low and high $f_m$‘s and a maximum for an intermediate $f_m$, Fig.5.10. The maximum particles velocity $\langle v_p \rangle \approx 1.5–2 \mu m/s$ is achieved at $f_m \approx 2–10$ Hz. This is exactly what we observe in the simulations for
Fig. 5.9. The calculated average flow velocity profile observed under modulated signal for different modulation frequencies $f_m$ (simulations are made by S. Tang).
Fig. 5.10. Average particles velocity $\langle v_p \rangle$ vs. $f_m$ for $f_c = 10$ kHz measured in the experiment. The line shows the simulated average backflow velocity $\langle v \rangle(f_m)$ at $z = 5 \mu m$. The inset shows the enlarged plot for $0 < f_m \leq 10$ (simulations of $\langle v \rangle(f_m)$ are made by S. Tang).

$\langle v \rangle(f_m)$, Fig. 5.9. Good agreement suggests that the prime mechanism of the particle migration in our experiments is the backflow effect.
5.5 Conclusions

We presented the experimental results on statics and dynamics of colloidal particles dispersed in nematic LC. The particles are big enough (microns) to cause dipole-like elastic distortions of the director around them. In the absence of electric field, the particles levitate in the bulk of the cell, as the gravity forces are opposed by repulsive forces between the particle and the bounding walls. By measuring the position of the levitating particle as a function of the cell thickness, we determine the elastic repulsive force between the particle and the rigid wall and find it to agree well with the predictions of the elastic theory by Poulin et al. [8].

In the absence of electric field, the elastic dipole is roughly horizontal and oriented along the direction of rubbing in the cell, in any of two available directions. When the field is applied, this symmetry is broken and the particles with opposite directions of the elastic dipole move to the opposite substrates. We discussed two possible mechanisms behind this effect, elastic trapping and dielectrophoretic effect. The elastic forces prevail at low fields (0.1 V/μm or less) while the dielectrophoretic forces dominate at higher fields.

The most dramatic effect of the applied electric field on the colloidal particles is through the backflow effect. The latter causes the particles movement along the bounding plates. Since the backflow is antisymmetric, the particles of opposite polarity of the elastic dipole are moving into opposite directions. Numerical simulations of the backflow profile are in a good agreement with experimental measurements of particles velocity. The effect of particles transport is similar to the one reported in Ref. [40] where
the particles motion and separation in isotropic fluid were achieved by creating a ratchet-type potential. In LCs these effects are further enriched by the elasticity-mediated interactions.

To conclude, the experiments demonstrate that the particles dynamics is controlled by director distortions and can be affected by the external fields. Overall, the colloidal dynamics in LCs demonstrate a rich variety of mechanisms and effects involved. Some of them are not explored yet. For example, one may expect a spatial separation of particles with different sizes under modulated signal (particles sorting effect). The mechanism behind this is that the bigger particles move slower than the smaller ones as a backflow velocity is $z$-dependent. Also, by reducing the thickness of the cell or increasing the diameter of the particles, one can strongly enhance particles interactions with at each other and thus study the phenomena such as jamming [41], Fig. 5.11; these studies are currently in progress.
Fig. 5.11. Preliminary results of clustering of the colloids observed in a planar cell under modulated signal. (a): Cartoon explaining aggregation dynamics of colloidal particles. (b): Polarization microscopy picture (experiment) showing aggregation of colloidal particles into clusters in a 10µm cell under modulated electric signal. Colloidal particles and electric signal are as described in Sections 5.2-5.4.

5.6 References


CHAPTER 6

Summary

6.1 Summary of the results and further developments

Nowadays LCs are at the core of modern technologies. New discoveries promise more LC-based applications to come either in a form of thin paper-like displays [1] or personal care products [2]. In the Dissertation we explored new approaches for electro-optical applications of LCs. We described new designs of LC-based devices as well as shed some light on relatively new phenomena, such as negative refraction and electrically-controlled dynamics of colloidal particles in LCs.

The main results obtained in this work are as follows:

- electrically tunable amphoteric (negative and positive) refraction was observed in nematic LC [3]. The very existence of negative refraction in LC has been proven. A tunable amphoteric refraction was realized by using a very simple geometry utilizing a nematic cell. Applications of the demonstrated phenomenon may include, e.g., beam steering devices. Future developments may include the utilizing LCs in heterogeneous materials, for example, photonics crystals [4], where one can expect to achieve a tunable amphoteric refraction in a certain spectral range.
- applicability of SmA materials for birefringent prisms useful in digital beam steerers [5-7] and dual-frequency nematic for fast achromatic polarization rotator [8] has been demonstrated. SmA-filled prisms are attractive in low-cost applications where one needs large apertures. Structural flexibility of LC director allows for non-trivial geometries of LC-based optical elements, such as SmA-filled blazed gratings and (possibly) curved geometries. We demonstrated that the mixtures of homologues of 4,4'-n-dialkylazoxybenzene produce SmA phases with a broad temperature range of SmA existence (up to 30°C for binary mixtures) with a relatively small number of residual defects, such as FCDs, and rather high transmission characteristics above 70% for both ordinary and extraordinary waves even when the SmA layer becomes thicker than 1 mm (no antireflection coatings were utilized). We determined the typical magnetic fields needed to remove director distortions around the mechanical inclusions and FCDs. Magnetic alignment is most effective when the material is aligned in the nematic phase and then cooled down to the SmA phase [7]. For DBDs we also developed a broadband polarization rotator based on two 20 µm thick 45° TNs with opposite handedness. Implementation of a high pretilt angle of n near the boundaries, dual-frequency feature of the LC material and overdriving technique allows one to significantly reduce the switching time of the rotator down to 2 ms. The rotator showed high transmission >87% in a wide spectral range 0.5 µm ≤ λ ≤ 2.7 µm and can be used in DBDs or in optical elements where fast rotation of the polarization state of multiband light is required.

- applicability of dual-frequency nematic for LC-based lens with a hole-patterned electrode structure, which focal length can be tuned by the electric field from negative to
positive values, has been demonstrated [9]. We achieved relatively fast response time about 0.4 sec during focus change for a 110 $\mu$m thick cell (compare with about 10 sec for conventional device of the same thickness); most of the director structure is switched within the first 50-100 ms. The optical power of the system with the aperture $D=300 \mu m$ can be continuously tuned from -520 to +400 diopters ($\lambda = 633 \text{ nm}$) in a double LC lens approach. Such a design can be used to achieve fast optical communication between multiple channels, for example, in microlens arrays, in beam steering or scanning devices or for fast non-mechanical zooming in miniature cameras and can be easily upscaled to the 1-5 millimeter-range aperture, see, for example, Ref. [10]. Further developments may also include a mitigation of electrically-induced backflow in both TN cells and tunable lenses for even more improved switching characteristics of the devices.

- behavior of colloidal particles dispersed in a nematic LC and surrounded by hyperbolic-type defects has been explored [11, 12]. We observed a number of interesting effects, such as levitation of particles in the bulk, selective movement of particles toward the opposite substrates according to the orientation of satellite defects and bidirectional motion controlled by electrically-induced backflow. By measuring the position of the levitating particle as a function of cell thickness, we determined the elastic repulsive forces between the particle and the rigid walls as well as discussed the elastic and dielectrophoretic forces among the possible mechanisms of the particle’s lift under applied field. We found that a bidirectional motion of colloidal particles is in a good agreement with a velocity and profile of the electrically-induced backflow. This fact confirms that the prime mechanism for particles transport is a backflow effect. The
observed phenomena open the possibility for electrically-driven particles manipulation in LCs. This may result in many practical applications, including particles sorting, electrically-driven LC microfluidic devices, development of LC-based electrophoretic displays, etc. The studied LC-based colloidal dispersions also offer an excellent model system to study viscoelastic properties of complex fluids, Brownian motion, interparticle interactions, hydrodynamics, pedestrian and traffic behavior, aggregation and jamming phenomena [13], etc. These studies are currently in progress.

6.2 References


2. See, for example, the website: http://www.pantene.com/en-US/product/blondefluxexpress_dailycolorenhanceshampoo.jspx.


