Liquid crystal based tunable photonic devices

Luo, Dan

2012

Luo, D. (2012). Liquid crystal based tunable photonic devices. Doctoral thesis, Nanyang Technological University, Singapore.

<https://hdl.handle.net/10356/53691>

<https://doi.org/10.32657/10356/53691>

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LIQUID CRYSTAL BASED TUNABLE PHOTONIC DEVICES

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A thesis submitted to the Nanyang Technological University
in partial fulfillment of the requirement for the degree of
Doctor of Philosophy

2012

Acknowledgements

I would like to take this opportunity to express my sincere gratitude to my supervisor Prof. Sun Xiaowei for his guidance and advices in the four years of my PhD studies. I have been benefited a lot from his enthusiasm for science, extensive expert knowledge, constant support and encouragement in my research. I would also like to thank Dr. Liu Yanjun, for the valuable comments and constructive suggestions he gave me during my process of research, and my thanks also go to Dr. Dai Haitao for the helpful discussion.

I am grateful to Prof. Ji Wei and Mr. Yang Hongzhi in National University of Singapore for their Nd:YAG laser system and useful discussion. I would like to thank for the help and the assistance of Prof. M. B. H. Breese and Mr. O. Y. Sheng in National University of Singapore. I would like to thank Dr. Yin Xijiang and Mr. Goh Kwee Hwang in Singpaore Polytechnic, for their help in carrying out some of the experiments and tests.

I would like to extend my gratitude to all the staffs and students at Nanoelectronics I, Network Technology Research Center, and *Luminous!* for their kind support and assistance. Special thanks to Prof. Shum Ping, Prof. H. V. Demir, and Prof. Sun Changqing for their insight views and fruitful discussions; to Dr. Tan Swee Tiam, Dr. Yang Yi, Dr. Wang Jianxiong, Dr. Divayana Yoga, Ms. Wang Ying, Ms. Ling Bo, Mr. Wang Ye, Mr. Iwan Sugihartono, Mr. Zhao Dewei, Mr. Du Qingguo and all the friends from the above three laboratories for their friendship, encouragement, inspirational discussions, and all the different kinds of assistance they provided to me.

Finally, I would like to express my deep appreciation to my parents and my wife for their continuous encouragement and support.

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Summary

Summary

Liquid crystals (LCs), which was firstly discovered by Austrian botanical physiologist Friedrich Reinitzer in 1888, are substances with a phase of matter whose properties between those of a solid crystal and those of a conventional liquid. The property of responding to external electric field and optical anisotropy makes liquid crystals an excellent candidate of tunable diffractive optical elements. Liquid crystals based diffractive optical elements combine the advantage of liquid crystals and diffractive optical elements (DOEs), and also broad the application range of such diffractive optical elements.

Holographic polymer-dispersed liquid crystals (H-PDLCs), which hold promise for many applications in optical communications, display, information storage and integrated optics, are formed by combining the holography and polymer-dispersed liquid crystals (PDLCs). Photonic crystals (PCs), which are attractive optical materials for controlling and manipulating the flow of light, have been studied for many years. With doped dye as the gain medium, laser can be generated from dye doped 1D and 2D H-PDLC defect free photonic crystals. As an organic material, H-PDLC is a good choice of dynamic mirrorless all organic tunable laser device.

In this dissertation, studies are focused on two parts: one is on developing new liquid crystals based diffractive optical elements, and another is on investigating lasing from two-dimensional holographic polymer dispersed liquid crystals photonic crystals.

For the first part, three different kinds of LC based diffractive optical elements were studied. They are electrically-switchable computer-generated

Summary

hologram (CGH) fabricated by a LC cell with polymethylmethacrylate (PMMA) film pattern; electrically switchable finite energy Airy beams generator fabricated by a LC cell with one electrode patterned with a binary-phase distribution; a polarization-dependent circular Dammann grating (CDG) fabricated an azo-dye-doped LC cell by applying simple multi-exposure photo-alignment process. The intrinsic properties such as electrically tunable characteristics, diffraction efficiency, or polarization dependent properties were demonstrated respectively for each diffractive optical element.

For the second part, lasing actions were demonstrated in two high-symmetry directions (ΓM and ΓK) of a two-dimensional hexagonal photonic crystal from dye-doped H-PDLC. The group velocity anomaly, which was peculiar to two- and three-dimensional photonic crystals, led to substantial enhancement of local fields in the two-dimensional photonic crystal with a low-index contrast. Besides, the mode dependent electrically tunable property of lasing for transverse electric (TE) and transverse magnetic (TM) modes, and the temperature dependent property of lasing along ΓM direction in TE polarization were also investigated in dye-doped two-dimensional hexagonal photonic crystal made of H-PDLC.

Chapter 1 Introduction

1.1 Motivation

1.1.1 Liquid Crystals

Introduction of Liquid Crystals

Liquid crystals (LCs), which were firstly discovered by Austrian botanical physiologist Friedrich Reinitzer in 1888, are substances with a phase of matter whose properties are between those of a solid crystal and those of a conventional liquid [1]. This phase is referred to as the liquid crystal phase coined by Otto Lehmann in 1889. Materials capable of forming liquid crystal phases are called mesogens.

Liquid crystals can be divided into three types: thermotropic LCs, lyotropic LCs, and polymeric LCs [2]. The thermotropic LCs exhibits a phase transition as temperature is changed while the lyotropic LCs exhibits phase transitions as a function of an appropriate concentration of materials is dissolved in some solvent. In polymeric LCs, rigid structures of polymer resemble the LC molecules with some orientational and positional order characteristic of liquid crystals. While all liquid crystals oppose molecularly anisotropy, the natures of this shape anisotropy are divided into two classes: calamitic liquid crystals with rod-like shape and discotic liquid crystals with disk-like shape. Typically, calamitic liquid crystal has elongated molecules and the director is defined as the direction of the long axis of the molecule [3].

Liquid Crystal Phase

The thermotropic liquid crystals are the most widely used and studied for their optical linear and nonlinear properties. There are three main phases of

thermotropic LCs: nematic phase, smectic phase, and cholesteric phase. The structure of the constituent LC molecules, also named the building blocks, determines the types of liquid crystals that can be observed. Depending on the nature of the building blocks and the external influence factors, such as temperature or solvents concentration, a wide variety of transition among liquid crystal phase could be observed. Two temperatures here are important to understand the phase of thermotropic liquid crystals: T_M is the melting temperature, bellowing which the materials are solid, crystalline and anisotropic, while $T_C > T_M$ is the clearing temperature, beyond which the materials become isotropic liquid. Between T_M and T_C , the materials are in the liquid crystals phase. Figure 1-1 shows schematic of three main phases of thermotropic liquid crystals. Other phases include blue phase and discotic phase etc.

Figure 1-1 Phases of thermotropic liquid crystals: (a) Nematic Phase; (b) Smectic Phase; and (c) Cholesteric Phase [3].

In a nematic phase, the molecules, on average, orient about a particular direction, called director, showing only a one-dimensional orientational order, without translational order. In a smectic phase, the LC molecules have aligned

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themselves in planar sheets. There are several sub-classifications in accordance with the positional and directional arrangement of the molecules, meaning that it has orientational and one-dimensional translational orders. Smectic A phase and smectic C phase are two common classes of smectic phase. For smectic A phase, the director of the molecules are along to layer normal. For smectic C phase, the molecules are arranged with random deviations tilted to the layer normal. The cholesteric phase, also called chiral nematic phase, is actually a nematic type exhibits a twisting of the molecules normal to the director with the molecular axis aligned along to the director. Blue phase is a special type, which appears in the temperature range between an isotropic phase and a cholesteric phase, possessing a repeated three-dimensional cubic structure of defects with lattice periods in the range of several hundred nanometers. There the blue phase reflects light in visible range, which is called Bragg reflections. Discotic phase is formed by disk-shaped mesogens orienting themselves in a layer-like pattern called discotic nematic phase.

Liquid Crystal Order Parameter

Liquid crystal molecules in the nematic phase are rod-like and easy to align along one direction. The unit director $\vec{n}$ can be defined to represent the ensemble average of the direction [3]. An order parameter is used to define the degree of orientational ordering in a liquid crystal. In a nematic liquid crystal, this order parameter is [4]:

$$S = \frac{1}{2} \langle 3 \cos^2 \theta - 1 \rangle, \quad (1.1)$$

where θ is the angle between the director and specific molecular long axis (shown in Figure 1-2), and the brackets represents the ensemble averaging. For

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perfect alignment, $S=1$; while for disappear of orientational order or totally random orientation, $S=0$. For most of nematic liquid crystal, the value of order parameter is between 0 and 1, where 0.5~0.7 is the typical range [3]. Also, the order parameter for thermotropic liquid crystals is strongly temperature dependent. The order parameter gradually decreases as the increase of temperature and will suddenly drop to zero while the temperature reaches the isotropic-nematic transition point, or so called clearing temperature. An example of the order parameter as a function of temperature is shown in Figure 1-3 as a curve, according to Maier-Saupe mean field theory [3].

Figure 1-2 Orientational order in nematic liquid crystal molecules. $\vec{n}$ represents the director. θ is the angle between the molecular long axis and the director [3].

Figure 1-3 Nematic liquid crystal order parameter varies as a function of temperature [3].

Anisotropy in Liquid Crystal

Due to the shape anisotropy of nematic liquid crystal, the physical properties of the liquid crystal such as dielectric constant, refractive index, and magnetic susceptibility. With an applied electric field on a nematic liquid crystal, the charge distribution in the molecules will be changed and thus leads to the dipole moments. For weak field limit, the induced polarization $\mathbf{P}$, generated by the electric field ($\mathbf{E}$) of the nematic liquid crystal is given by [3]:

$$\mathbf{P} = \epsilon_0 \chi \mathbf{E}, \quad (1.2)$$

where $\epsilon_0 = 8.86 \times 10^{-12} \text{ C}^2 / \text{N.m}^2$ is the permittivity in free space, χ is the material's macroscopic susceptibility tensor mainly related to the orientational order [3]. Because of the shape anisotropy of the nematic liquid crystal molecule, the susceptibility differ in value in the case of applying electric field: parallel ($\chi_{\parallel}$) and perpendicular ($\chi_{\perp}$) to the nematic director. From the defining

of electric displacement field $\mathbf{D} = \epsilon_0 \mathbf{E} + \mathbf{P} = \epsilon \mathbf{E}$, we have $\epsilon = \epsilon_0(1 + \chi)$. The dielectric anisotropy is defined as:

$$\Delta\epsilon = \epsilon_{\parallel} - \epsilon_{\perp}, \quad (1.3)$$

where $\epsilon_{\parallel}$, and $\epsilon_{\perp}$ are the dielectric permittivity parallel and perpendicular to the LC molecular symmetry axis, respectively [3]. The induced dipole moment will play a role to reorient the nematic director to be parallel to the applied electric field when $\Delta\epsilon > 0$, and to be perpendicular to the applied electric field when $\Delta\epsilon < 0$. The value of $\Delta\epsilon$ is determined by the nematic LC chemical structure.

The dielectric constant permittivity of liquid crystal is dependent to the frequency. In optical regime, the ordinary refractive index n_o ($n_{\perp}$), corresponds to the light propagating in the liquid crystal with electric field polarization perpendicular to the director, while the extraordinary refractive index n_e ($n_{\parallel}$) corresponds to the light propagating in the liquid crystal with electric field polarization parallel to the director. The birefringence or optical anisotropy is given by:

$$\Delta n = n_e - n_o, \quad (1.4)$$

where we have the relationship of $\epsilon_{\parallel} = \epsilon_0 n_e^2$ and $\epsilon_{\perp} = \epsilon_0 n_o^2$. For $n_o < n_e$, LC is of positive birefringence; for $n_o > n_e$, LC is of negative birefringence.

Figure 1-4 (a) Dielectric anisotropy and (b) Optical anisotropy of a uniaxial nematic liquid crystal [3].

For light propagating with an incident angle of θ to the optical axis of liquid crystal, two refractive indices are seen by light with two modes of propagation. One mode is called ordinary wave with its polarization direction perpendicular to the plane formed by the optic axis and the wave vector, seeing a refractive index of n_o (independent of the incident angle θ). The other mode is called extraordinary wave with its polarization direction parallel to the plane formed by the optic axis and the wave vector, seeing an effective refractive index of n_{eff} (dependent of θ). The effective refractive index can be written as

$$n_{eff}(\theta) = \left[\frac{\cos^2 \theta}{n_o^2} + \frac{\sin^2 \theta}{n_e^2} \right]^{-\frac{1}{2}}. \quad (1.5)$$

The optical anisotropy is positive for nematic liquid crystals, and negative optical anisotropy can be obtained in discotic materials [1]. The dielectric and optical anisotropy of a uniaxial nematic liquid crystal is indicated in Figure 1-4. Similarly the optical anisotropy is a function of temperature and approximately proportional to the order parameter S [5]. Figure 1-5 shows the

indices of refraction as a function of temperature. We can observe that, the optical anisotropy Δn narrows as the increase of temperature and finally jumps to zero in the isotropic phase.

Figure 1-5 A general trend of refractive index as a function of temperature for typical nematic liquid crystals, where T_C is the clearing temperature of LC, and n_{iso} is index of LC in isotropic phase [3].

Elastic Continuum Theory of Liquid Crystal

Without constraint, the nematic liquid crystal will align random in most of cases interested. Usually some kinds of physical constraint are used to align the liquid crystals through the surface alignment effect, which works through influencing the director orientation of liquid crystals. In most cases, distortions of the director from uniform alignment exist but happen over microscopic lengths [3]. The small distortion leads to a safety assume of that the order parameter can be treated to be constant. Considering only the elastic free energy of liquid crystals, and based on the assumption above, a continuum theory has been developed [4]. In an equilibrium state, the director of nematic liquid crystals is assumed to be constant everywhere. When this equilibrium state is perturbed, means in non-equilibrium state, the nematic liquid crystal will

undergo deformation just like any solid. Three basic deformations types of nematic liquid crystals system are called splay, twist and bend deformation, respectively, as illustrated in Figure 1-6. According to Oseen-Frank theory, the total distortion free energy density in non-equilibrium state can be written as [8, 9]:

$$F = \frac{1}{2} K_{11} (\nabla \cdot \mathbf{n})^2 + \frac{1}{2} K_{22} (\mathbf{n} \cdot \nabla \times \mathbf{n})^2 + \frac{1}{2} K_{33} (\mathbf{n} \times \nabla \times \mathbf{n})^2, \quad (1.6)$$

where K_{11} , K_{22} and K_{33} are Frank splay, twist and bend elastic constants, respectively.

Figure 1-6 (a) Equilibrium state, (b) Splay elastic deformation, (c) Twist deformation, and (d) Bend deformation in nematic liquid crystals [3].

Liquid Crystal Alignment

In a liquid crystal sample without alignment, the range order is absent in nematic liquid crystals, leading to a strongly light scatters of the bulk material. The scattering light in nematic liquid crystal usually has 10^6 times large intensity than that in traditional isotropic liquids [3]. It is necessary to properly

align the nematic liquid crystals to obtain good optical performance when applying nematic liquid crystals as a practical device and component.

Homogeneous and homeotropic alignment are the two common categories of liquid crystal alignment geometries, the alignment of liquid crystals in both structures are shown in Figure 1-7. In homogeneous alignment structure the nematic director is aligned parallel to the substrate surface while in homeotropic alignment the nematic director is aligned perpendicular to the surface. Besides above two kinds of alignments, another alignment called twisted alignment is also used especially in LC display industry. A popular used 90° twisted nematic (TN) configuration can be created by using two pieces of homogenously rubbed glass substrates with orthogonally arrangement. The TN cell can operate in normally white and normally black mode depending on the different setting of polarizers' directions used.

Figure 1-7 Homogeneous and homeotropic alignment geometries [3].

These alignments can be achieved by mechanical, chemical or optical treatments of the substrate surface. In 1911, Mauguin introduced the process of

rubbed polymer layers to achieve homogenous alignments of nematic liquid crystals [8]. Microgroove structure, which can be generated through unidirectional polishing, tangential evaporation, was also found to be used to align liquid crystals due to the formation of relief grating structure on the substrate through a mechanical rubbing on the substrate [9, 10]. Besides, optical alignment methods were introduced in order to avoid contaminating impurities or electrostatic charges during the rubbing process [11-13]. Other alignment methods include twisted and super twisted alignment, which are often found their applications in liquid crystal display (LCD) technology.

1.1.2 Photopolymerizable Polymers

Polymer and Photopolymerization

Polymers are a class of soft condensed matter usually found in our normal life. The typical polymer molecule is formed by a chain of small species named monomer. Monomers are stable molecules that can be combined to synthesize polymers. The number of reactive sites on an individual monomer is known as its functionality. Depending on the number of functionalities of the monomer, different morphologies of polymer networks can be formed [14]. A linear chain is formed by di-functional monomers. A branched polymer is formed if higher functional monomers are involved. The branches start to connect to one another in three dimensions if the functionality is even higher where the polymer forms a crosslinked network. Figure 1-8 shows the three typical polymer structures.

Figure 1-8 Polymer structure of (a) linear chain, (b) branched chain, (c) crosslinked network. The circle represents each monomer unit [3].

Condensation and free radical polymerization are two basic types of polymerization reactions. In condensation polymerization, the monomers combine with each other to form polymer chains with a by-product of water. In free radical polymerization, the double bond between carbon atoms will be broken and leave one unshared electron on each carbon atom, leading to the carbon with an unpaired electron which is very reactive and known as a free radical. Two free radicals can form a new covalent bond linking between two monomers.

Photopolymerization is a kind of polymerization process induced by light exposure. Due to the flexibility and controllability of light's properties, such as wavelength, intensity, and polarization state, this kind of photopolymerization are popular in industry and research fields. One example of photopolymer is the photoresist, which is one of the most important processing materials in electronic and display industries.

Polymer Stabilized Liquid Crystals

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Due to the electro-optical property of liquid crystal and the morphologies of polymer, liquid crystal/polymer composites can be used in many interesting devices. Depending on the different concentration of polymer within the LC/Polymer composites, two typical types of LC/Polymer composites, polymer-stabilized liquid crystals (PSLC) and polymer dispersed liquid crystals (PDLC) can be formed.

In PSLC, the polymer concentration is quite low and typically around 5wt%. The liquid crystals materials of PSLC system form the matrix while the crosslinked polymers disperse in the liquid crystal fluid. Generally, like the liquid crystal, the most of practical PSLC devices need the pre-treatment of substrate, e.g. glass surface, to align the orientation of liquid crystals molecules inside of PSLC. The phase of polymer and liquid crystal can be separated by UV light source, where the mixture of monomers and liquid crystals getting through the process of photopolymerization. The finally formed polymer network can be isotropic or anisotropic depending on the specific order of liquid crystal and the properties of monomers. The advantage of PSLC comparing to the PDLC we will introduced following is the low operation voltage while the disadvantage is the low response time in electric field.

Polymer Dispersed Liquid Crystals

Polymer dispersed liquid crystals is composed of discrete liquid crystals droplet with micron-size randomly dispersed throughout a rigid polymer matrix. The typically concentration of polymer in PDLC is larger than 20wt%, comparing to a typically 5wt% polymer concentration in PSLC.

PDLCs can be prepared by two different methods: phase separation [15] and encapsulation [16]. Phase separation methods start with a homogenous

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mixture, where liquid crystal droplets component is phase separated from polymer. The phase separation can be triggered using several ways including solvent induced phase separation (SIPS), thermally induced phase separation (TIPS) and polymerization induced phase separation (PIPS). On the other hand, the encapsulation method, also called nematic curvilinear aligned phase, is formed by the dissolving liquid crystal into a water-borne polymer to generating an emulsion, which subsequently being coated on a substrate surface. Before the film is coated and dried, the LC droplets can be created in the process of emulsification.

PDLCs are typically filled between transparent conducting Indium Tin Oxide (ITO) coated glass substrates. The LC droplets size formed within PDLCs can be influenced by environment temperature, exposure light intensity, and exposure duration time of light. Two typical nematic director configurations in spherical cavities of LC droplets in PDLCs are called homogeneous parallel anchoring, leading to a bipolar and aligned bipolar (in external electric field) configuration, and homeotropic anchoring, leading to a radial and axial (in external electric field) configuration, as shown in Figure 1-9.

Figure 1-9 Nematic director configuration of LC droplets in PDLC as (a) bipolar, (b) aligned bipolar, (c) radial, and (d) axial configuration, respectively [3].

In most of interest cases, homogenous anchoring is preferred, thus a bipolar configuration is formed. Without external electric field applied, the bipolar axes are dispersed in polymer matrix randomly. The refractive mismatch between the liquid crystals droplet and polymer matrix will lead to a significant incoherently light scattering, making an opaque state. As voltage is applied and higher than the threshold, the bipolar LC droplets will be aligned with the director of droplet symmetry axes along the external electric field direction. By properly choosing the index of polymer (n_p) to be equal to the ordinary index (n_o) of LC in PDLC, light propagation through PDLC along the optical axis will not experience a refractive index mismatch, thus the PDLC material appears a transparent state to light. The schematic of opaque and

transparent state of PDLC without and with external electric field are shown in Figure 1-10.

Figure 1-10 Schematic (a) opaque state, and (b) transparent state of PDLC without and with external electric field [3].

Holography Interference

Holography is a process of recording the complete amplitude and relative phase information of interference of light waves in an optical active medium and consequently a real three-dimensional image can be reconstructed [17]. It has long been a powerful and proven tool useful in many areas of optics, precision measurement, and information processing/storage. The idea of holographic imaging was firstly proposed by Gabor in 1948 [18].

Before specific H-PDLCs structures, we firstly introduce the basic principle of holography lithography or optical interference, which is a method based on the superposition of two or more coherent light waves. Assume that the complex amplitude electric field vector of the m th plane wave is [19]:

$$\mathbf{E}_m(\mathbf{r}) = E_m \exp(i\mathbf{K}_m \cdot \mathbf{r} + i\delta_m) \mathbf{e}_m, \quad (1.7)$$

where E_m is the real amplitude, $\mathbf{K}_m$ is the propagation vector, $\mathbf{r} = (x, y, z)^T$ is the position vector, δ_m is the initial phase, and $\mathbf{e}_m$ is a unit vector in the plane wave's polarization direction. The intensity distribution of the interference field can be defined as:

$$I(\mathbf{r}) = \mathbf{E}_{tot}^* \cdot \mathbf{E}_{tot} = \Re \left(\sum_{l,m=1}^N \mathbf{E}_l^* \cdot \mathbf{E}_m \exp[i(\mathbf{K}_l - \mathbf{K}_m) \cdot \mathbf{r} + i(\delta_l - \delta_m)] \right), \quad (1.8)$$

where $\mathbf{E}_{tot} = \mathbf{E}_1 + \mathbf{E}_2 + \dots + \mathbf{E}_N$, N is the total number of the interference plane waves, and $\Re[\dots]$ denotes the real part of the argument.

Holographic Polymer Dispersed Liquid Crystals

Holographic polymer-dispersed liquid crystals (H-PDLCs) are formed by combining the holography and PDLCs. Comparing to PDLC, the H-PDLCs have two major distinguish characters: Firstly, the LC droplets in H-PDLCs are confined to well-defined planes as one dimensional (1D), two dimensional (2D) or three dimensional (3D) structures, e.g. photonic crystals, where the droplets are randomly dispersed throughout the polymer in PDLC; Secondly, the size of liquid crystal droplets (typically several hundred nanometers) in H-PDLCs are significantly smaller than that of PDLCs (typically micrometer-sized). Other features of H-PDLCs include its true volume hologram character, high index modulation, unique anisotropic nature, and electro-optical behaviour.

In 1993, Sutherland [20] et al fabricated a PDLC Bragg grating by holography, after that, a lot of research works concerning about H-PDLC grating [21] have been reported, including studies on polymer networks [22], two-photon induced polymerization renditions [23] and total internal reflection

configuration [24] of H-PDLC grating. Also, there are many basic studied on polarization properties and electro-optical switching behaviour of H-PDLC grating [25, 26], formation of high-resolution H-PDLC grating [27], electrical, thermal and optical dynamic properties [28] and anisotropic diffraction H-PDLC gratings [29].

H-PDLCs grating can be formed through exposing of LC/Polymer mixture samples on two beam interference pattern and can be divided into two groups: transmission and reflection H-PDLC gratings. The holographic exposure initiates Photopolymerization, which leads to a diffusion of liquid crystal to the dark regions of the interference pattern and monomer diffusion into the bright regions of the interference pattern. The monomer subsequently polymerizes as polymer matrix and resulting an alternating LC rich and polymer rich regions in grating periodic structure [30]. The LC droplets of H-PDLCs are influenced by factors of light exposure time and intensity, and the concentration ratio of LCs and monomers etc. Figure 1-11 shows the schematic configurations of transmission and reflection H-PDLC gratings produced by two interference light beams.

Figure 1-11 Schematic configurations of (a) transmission and (b) reflection H-PDLC grating produced by two interference light beams.

The generated interference period of the grating (Λ) with two beams can be expressed as:

$$\Lambda = \frac{\lambda}{2n\sin\theta} \quad (1.9)$$

where λ is the light beam wavelength, n is the average refractive index of LC/Polymer mixture, and θ is equal to the half angle between two incident beam inside the LC/Polymer mixture. Polarization optimization is usually straightforward as parallel polarizations leads to the highest interference contrast, and cross polarizations leads to no interference contrast, and other polarizations leads to something in between.

1.1.3 Nematic Liquid Crystals Based Tunable Diffractive Optical Elements

Diffractive optics plays an important role in a variety application in modern optics. Diffractive optical elements (DOEs) are useful tools based on the principle of diffractive optics with applications in three-dimensional displays, holography, microlens arrays, high efficiency gratings, beam splitting and shaping devices [31], optical computing architectures and photonic switching [32], and laser machining acting as welding, cutting or drilling tools [33]. Comparing to conventional bulk optics, DOEs have a lot of intrigue advantages including functional versatility, small size and weight, high efficiency, the ability to be integrated with other devices, smaller features, and low cost fabrication, due to its planar nature. These special properties make DOEs to be useful in many applications as photonic devices. Fabrication methods of DOEs include microphotolithography, electron beam writing, laser beam writing, and imprinting etc.

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Conventional materials used in diffractive optical elements are usually non-tunable materials such as glass, PMMA, and photo refractive crystals. Liquid crystals based diffractive optical elements combine the advantage of liquid crystals and diffractive optical elements, and also as broad the application range of such diffractive optical elements. The principle of nematic liquid crystals based DOEs are due to the phase modulation between different phase zones formed by liquid crystals with different orientation states, which can be artificially controlled by external parameters, such as electric field, and temperature directly, or realized through influencing different dye materials doped in nematic liquid crystals.

The property of responding to external electric field and optical anisotropy make liquid crystals to be an excellent candidate of tunable diffractive optical elements. Recently, a lot of research works have been reported on useful diffractive optical device such as Fresnel zone lens [34-41], optical vortex [42, 43], fractal zone plates and fractal photon sieves [44], diffractive lens [45], using many different methods including nematic liquid crystals, twisted nematic liquid crystal, azo-dye doped liquid crystal, liquid crystal polymer-separated composite, polymer-dispersed liquid crystals, and polymer-stabilized liquid crystals etc. The replacement of tunable material of nematic liquid crystals to index-fixed materials as mentioned above provides significantly improved flexibility and controllability to the liquid crystals based diffractive optical elements, making the LC based DOEs as a promising application-widely optical-electronic devices with the advantage such as low operating voltage and fast response time.

1.1.4 Lasing from Two-Dimensional Holographic Polymer Dispersed Liquid Crystals Photonic Crystals

Photonic crystals (PCs) are periodic optical nano-structure designed to control the motion of photon. They are similar to semiconductors with periodic structure in atomic scale. Generally, photonic crystals contain regularly repeating internal regions of high and low dielectric constant. Photonic crystals are attractive optical materials for controlling and manipulating the flow of light. As the development of theoretical simulation and experimental fabrication technique, photonic crystals are found to be used in linear optics, nonlinear optics and quantum optics. Many novel devices based on photonic crystals have been reported: such as high Q-value micro cavity [46], photon frequency bandgap planar antenna [47], high efficiency light emitting diode [48], linear and nonlinear optical waveguide [49-51], optical filter in communication [52], photonic crystals fibers [53, 54].

As a good choice of miniature laser, laser based on photonic crystals fabricated by inorganic material, such as semiconductors, arouse a lot of concerns in the recent year. The miniature laser is a good candidate for on-chip sources in integrated photonics. Depending on the photonic crystals structure where laser generated from, lasing from photonic crystals can be divided into two types: one is the most popular and important type of photonic crystal lasers is laser generated from bandgap defect-mode of photonic crystal with defect as cavity, where lasing action arise from the resonant cavity modes [55-57]. Another type of photonic crystal laser is generated from photonic crystal without defect, where no physical cavity exists and operation of laser is based on enhancement of the optical density of state due to small group velocity [58].

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In the second type of laser from defect free photonic crystal, the mechanism behind the laser from 1D photonic crystal grating is similar to the conventional 1D distributed feedback lasers, while the mechanism behind the laser from 2D photonic crystal depends is the high density of state due to low group velocity at band edge [59-62].

Beside inorganic material, organic material H-PDLCs is a good candidate to fabricate different kinds of 1-D, 2-D, and 3-D photonic crystals by using properly two-beam, three-beam, and four-beam holography interference technique. Although the index contrast of photonic crystal fabricated by H-PDLC is quite small, laser action also can be realized through the distributed feedback mechanism for 1D PC and group velocity anomaly for 2D PC. With doped dye as gain medium, laser can be generated from dye doped 1D and 2D H-PDLC photonic crystals, where a second type of laser from defect free photonic crystal can be oscillated. Comparing to the semiconductor materials, the organic/active materials (H-PDLC) have several advantages such as: easy fabrication process, fast response, large fabrication area, electrically tunable, and temperature dependent characteristics. Organic material such as H-PDLC is a good choice of dynamic mirrorless all organic tunable laser device.

1.2 Objective

This thesis addressed the design and fabrication of liquid crystal based tunable photonic device. Two major parts are included in our investigation field. The first part discusses about new applications of liquid crystals on diffractive optical elements, and their electric and optical performances will be investigated. The second part discusses the mechanism behind the laser devices based on 2D H-PDLC photonic crystal. The directional laser from 2D H-PDLC

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hexagonal structure will be investigated and the electrically tunable and temperature dependent property of laser will also be discussed.

Our first goal is to extend the applications of tunable LC based diffractive optical elements. Three useful LC based tunable DOEs: computer generated hologram (CGH), Airy beam generator, and circular Dammann grating (CDG), will be demonstrated using nematic liquid crystal with PMMA film, nematic liquid crystal, and azo-dye doped nematic liquid crystal, respectively. The application range and potential of those devices will be enhanced as combining with powerful tunable material of liquid crystal. Electric and optical characteristics of those three diffractive optical elements, such as diffractive efficiency of reconstructed image, electro-optical characteristics of devices, or polarization property, will be discussed in detail separately.

Our second goal is to generate and study the special mechanism of directional lasing in 2D H-PDLC photonic crystals devices. Firstly, we will discuss how to fabricate 2D photonic crystals with hexagonal structure using H-PDLC with special designed prism, where properly controlling the interference laser light become critical. Secondly, by doping dye with suitable absorption and emission spectrum into those fabricated 2D H-PDLC photonic crystals, we will investigate the lasing phenomenon due to group velocity anomaly, which is specially in 2D and 3D photonic crystals and different from the 1D photonic crystal grating laser where distributed feedback mechanism is the underlying reason. Thirdly, the electrical and temperature dependent characteristics of lasing from 2D H-PDLC PCs for two different polarization lights will also be discussed in detail. The tunable laser generated has wide potential application.

1.3 Major contributions

- An electrically switchable computer generated hologram using a liquid crystal cell with proton beam patterned polymethylmethacrylate (PMMA) film has been investigated. This device is promising in adaptive optical element with relatively low operating voltage and high resolution. The diffraction efficiency and electro-optical performance of CGH has also been investigated.
- A pair of electrically switchable two-dimensional finite energy airy beams has been generated using a liquid crystals cell with binary-phase patterned electrode. These Airy beams are electrically switchable, which can be generated or erased with the change of applied electric field. The diffraction-free and transverse acceleration dynamics of the Airy beams were verified. This kind of binary-phase element based Airy beam device has the advantages of low cost, easy fabrication process, and electrical tunability, potentially useful in adaptive optics.
- A broad band polarization-dependent circular Dammann grating has been fabricated from an azo-dye-doped nematic liquid crystal cell. A binary phase liquid crystal two-order CDG zone plate consisted of an odd zone with twisted nematic (TN) LC structure and an even zone with homogenous LC structure was fabricated through a multi-exposure photo-alignment process. The CDG based on azo-dye-doped LC can be operated in a broad wavelength range due to the waveguide property of the TN LC structure.

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- Lasing actions along both ΓM and ΓK directions from a DCM dye-doped 2D hexagonal H-PDLC PC has been investigated. The group velocity anomaly is the underlying mechanism for the substantial light amplification in our 2D H-PDLC PCs with the low-index contrast. The big difference on the laser threshold along ΓM and ΓK directions is explained through the proportional relationship of the laser oscillation threshold and the group velocity of the corresponding band.
- The electrically tunable properties of lasing with multi-peaks from a dye-doped 2D H-PDLC PC with hexagonal lattice has been demonstrated. Over 10 nm red-shifting of the lasing peaks are obtained in non-polarization spectra when the applied voltage is increased up to 40 V. The effect of the applied electric field on the effective refractive index of LC are, however, also different for the TE and TM modes, which leads to an opposite band shifting and eventually results in the splitting of the lasing spectra between TE and TM modes.
- The temperature tunable properties of lasing from a dye-doped 2D H-PDLC PC with hexagonal lattice along ΓM direction have been investigated. When the temperature increases from 25 to 45°C, a red-shift of lasing peaks has been observed. The decrease of LC droplets' anisotropy with the increased temperature for TE polarization is the major underlying reason for downward movement of photonic band, which eventually leads to the red-shift of lasing peaks.

1.4 Organization

This thesis consists of 7 chapters.

Chapter 1 starts from the introduction of liquid crystals and holographic polymer dispersed liquid crystal with their basic property and characteristics, followed by the motivation and the objective of this thesis, finally the major contribution of this thesis has also been discussed.

Chapter 2 introduces a liquid crystals based computer generated hologram with its electrically switchable properties. The computer generated hologram was fabricated on a PMMA substrate using proton beam writing technique.

Chapter 3 reports the realization of finite energy Airy beam through a liquid crystal based binary cubic phase distribution. The electrically tunable property is investigated for such device. The diffraction-free and transverse acceleration dynamics of the Airy beams is verified. This kind of binary-phase element based Airy beam device has the advantages of low cost, easy fabrication process, and electrical tunability, potentially useful in adaptive optics.

Chapter 4 investigated a polarization-dependent circular Dammann grating diffraction from an azo-dye-doped LC cell. A simple multi-exposure photo-aligned process was used to fabricate a binary phase liquid crystal CDG zone plate that consisted of an even zone with homogenous LC structure and an odd zone with 60° and 55° TN LC structure. The polarization dependent property is achieved by rotating the direction of analyser behind the LC sample. The CDG diffraction from our azo-dye-doped LC cell can be generated by lasers within a broad wavelength band, which expands possible applications.

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Chapter 5 demonstrated lasing actions along both ΓM and ΓK directions from a DCM dye-doped 2D hexagonal H-PDLC PC. The 2D photonic crystal with hexagonal lattice structure was fabricated by three beam holography interference. The simulation result obtained from plane wave method is also given to compare with experimental result. The group velocity anomaly is used to explain the behind mechanism of lasing action in 2D H-PDLC photonic crystals.

Chapter 6 conducts an investigation on the electrically and temperature dependent characteristics on lasing from 2D hexagonal H-PDLC PC, which is discussed in chapter 5. The different effect of the applied electric field on the effective refractive index of LC on TE and TM modes is discussed. A splitting of the lasing spectra between TE and TM modes is obtained and explained from the opposite moving of photonic band structure. Also, the temperature tunable properties of lasing from a dye-doped 2D H-PDLC PC with hexagonal lattice along ΓM direction is presented. When the temperature increased from 25 to 45°C, a red-shift of lasing peaks has been observed. The decrease of LC droplets' anisotropy with the increased temperature for TE polarization is the major underlying reason for downward movement of photonic band, which eventually leads to the red-shift of lasing peaks.

Chapter 7 discusses the conclusion and future works of this thesis.

Chapter 2 Liquid Crystals Based Electrically Switchable Computer Generated Hologram

2.1 Introduction

2.1.1 Fourier transform

Fourier transform (F.T.) is an expression of the original function as a sum of other more elementary functions. The Fourier decomposition is useful in optics because the exponential functions behave in a simple way. Here we chose x, y as real space co-ordinates and ξ, η as frequency space co-ordinates. Given an arbitrary function $u(x)$, it can be decomposed as [63]:

$$\text{1-D Forward F.T.: } U(\xi) = \int_{-\infty}^{\infty} u(x) e^{-2\pi i \xi x} dx \quad (2.1)$$

$$\text{1-D Inverse F.T.: } u(x) = \int_{-\infty}^{\infty} U(\xi) e^{+2\pi i \xi x} d\xi \quad (2.2)$$

Those expressions can be easily extended to two-dimensional case. Since the exponential functions are separable, we can express the argument as a scalar product of two-dimensional vectors [63].

$$\text{2-D Forward F. T.: } U(\xi, \eta) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} u(x, y) e^{-2\pi i (\xi x + \eta y)} dx dy \quad (2.3)$$

$$\text{2-D Inverse F. T.: } u(x, y) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} U(\xi, \eta) e^{+2\pi i (\xi x + \eta y)} d\xi d\eta \quad (2.4)$$

2.1.2 Discrete Fourier Transform and Fast Fourier Transform

As the continuous Fourier transform of functions have been shown above, the actually calculation of Fourier transform is usually finished by a

Chapter 2 Liquid Crystals Based Electrically Switchable Computer Generated Hologram

discrete-to-discrete transformation using so called discrete Fourier transform (DFT). The input function used in DFT is discrete and whose non-zero values have a limited duration. Two dimensional DFT of periodic objects consisting of regular arrays of points can be expressed as [64]:

Forward DFT:

$$U_{jk} = \frac{1}{\sqrt{M}} \frac{1}{\sqrt{N}} \sum_{m=-\frac{M}{2}}^{\frac{M}{2}-1} \sum_{n=-\frac{N}{2}}^{\frac{N}{2}-1} u_{mn} e^{-2\pi i(\frac{jm}{M} + \frac{kn}{N})}, \quad j = -\frac{M}{2}, \dots, \frac{M}{2}-1; k = -\frac{N}{2}, \dots, \frac{N}{2}-1 \quad (2.5)$$

Inverse DFT:

$$u_{mn} = \frac{1}{\sqrt{M}} \frac{1}{\sqrt{N}} \sum_{j=-\frac{M}{2}}^{\frac{M}{2}-1} \sum_{k=-\frac{N}{2}}^{\frac{N}{2}-1} U_{jk} e^{2\pi i(\frac{jm}{M} + \frac{kn}{N})}, \quad m = -\frac{M}{2}, \dots, \frac{M}{2}-1; n = -\frac{N}{2}, \dots, \frac{N}{2}-1 \quad (2.6)$$

A fast Fourier transform (FFT) is an efficient algorithm to compute the DFT and inverse DFT in computer. As a DFT decomposes a sequence of values into components of different frequencies, the computing is too slow to be practical as an FFT can significant reduce the practical calculation time.

2.1.3 Computer Generated Hologram

The word “hologram” is obtained from Greek words with meaning of “whole recording”, which distinguishes and tells itself from the photograph. The hologram records not only the amplitude of wave but also the phase information of wave at each point on its surface while a photograph records only the intensity of incident wave. In the publication of Gabor in 1948 [65], the author firstly created the conception of holography wave-front reconstruction and the so called on axis interfereometric hologram was introduced. As the holograms can be generated by computer, the name of

Chapter 2 Liquid Crystals Based Electrically Switchable Computer Generated Hologram

computer generated holograms (CGHs) become a proper description. In 1966, Lohmann and Brown [66] invented the binary computer generated hologram, where the powerful calculation ability computer was combined with that newly invented fast Fourier transform algorithm using detour phase to create holograms with transmittance of only zero and one across their surfaces.

The computer-generated holograms have a lot of good properties as an object requires non-real-exist and an ideal wavefront can be computed based on the diffraction theory and encoded into a tangible hologram. Computer-generated holograms are useful DOE for wave front manipulation [67], optical element testing [68], optical information processing [69, 70] and three-dimension display [71]. Various materials have been used to record CGH such as photorefractive materials including BaTiO₃ crystal [72], Fe doped LiNbO₃ crystal [73], and even bacteriorhodopsin [74]. CGH devices based on liquid crystal (LC) spatial light modulators (SLMs) have also been reported [75-77]. Several methods were proposed to code holograms such as the detour phase hologram [78], the synthetic binary amplitude holograms [79], the direct binary search [80], computer generated holograms with pulse density modulation [81], and iterative quantization of digital amplitude holograms [82]. The method achieved the best practical result in coding of computer holograms is the iterative Fourier transform algorithm (IFTA) [83], in relation to the phase retrieval problem.

In this thesis, we are not going to discuss the details of different complicated methods used to calculate CGHs, since the points are how to combine liquid crystals with CGHs and corresponding characteristic of those tunable devices. Here we only simply introduce the process of iterative Fourier

transform algorithm as shown in Figure 2-1. Taking 2D CGHs as an example, the process of iterative Fourier transform algorithm starts from the desired intensity distribution B_0 in image plane with random phase distribution as input parameter. An inverse fast Fourier transform (inverse FFT) is used to transfer the function formed in image plane of $\bar{F}=B_0F|F^{-1}|$ to an expression in CGH plane of $W=|A|\exp(i\theta)$. Next, by combining the amplitude of A_0 and the expression W , a distribution function $\bar{W}=A_0W|W^{-1}|$ has been generated, and used as the source of fast Fourier transform (FFT), which leads to a distribution of $F=|B|\exp(i\varphi)$ in image plane. We finish one round of iterative Fourier transform and the amplitude with phase distribution of F . Because the B_0 is the desired amplitude distribution in image plane, if the amplitude of F is not equal to B_0 , a new $\bar{F}$ will be generated with its amplitude corrected by B_0 . The process will repeat till certain error criterion about the difference between $|B|$ and B_0 satisfies. If the CGH is binary type, certain criterion will be used to create the binary amplitude from continuous amplitude.

Figure 2-1 Schematic process of iterative Fourier transform algorithm.

2.2 Experiment and Simulation

2.2.1 Experimental Process of CGH Using Liquid Crystals Cell with PMMA pattern

Previously, switchable CGH using polymer-dispersed liquid crystals (PDLC) [84, 85] and LC cell with patterned electrode are report in our group [86]. The phase shift can be obtained by reorientating LC directors in PDLC or LC cell when an external voltage is applied. Here, we investigate a CGH fabricated by a liquid crystal cell with a proton beam patterned polymethylmethacrylate (PMMA) substrate.

PMMA is a transparent thermoplastic which is used as light or shatter-resistant as an alternative of glass. It is the synthetic polymer of methyl mechacrylate. This kind of material was firstly invented in 1928 and was brought to market in 1933. The properties of PMMA include low cost, moderate, easy handing, and simple processing.

The phase distribution of the CGH was calculated by the conventional iterative Fourier transform algorithm [87]. The letters “NTU” was encoded into a CGH with 128×128 binary-phase-only pixels. Figure 2-3(a) shows the phase distribution of the CGH, where the black and white regions represent phase value of 0 and π respectively. Figure 2-3 (b) shows the simulated reconstructed image of the CGH. A focused 2 MeV proton beam (p-beam) was used to write the phase distribution of the CGH directly into a 5 μm thickness PMMA film, which was spin-coated on an indium-tin-oxide (ITO)-coated glass substrate. The fabrication was completed by the Centre for Ion Beam Applications (CIBA) at the National University of Singapore [88].

Proton beam (p-beam) writing is a new direct-writing process that uses a focused beam of MeV protons to pattern resist material (e.g. PMMA) at nano-dimensions. The primary mechanisms for producing structures in resist materials is, in general, bond scissioning in positive resists such as PMMA, or cross-linking in negative resists such as SU-8. In positive resists the damaged regions are removed by chemical development to produce structures, whereas in negative resists the development procedures remove the undamaged resist leaving the cross-linked structures behind. In p-beam writing the damage is caused by proton-induced secondary electrons [89].

Figure 2-2 Schematic of the p-beam writing facility at CIBA. MeV protons are produced in a proton accelerator, and a demagnified image of the beam transmitted through an object aperture is focused onto the substrate material (resist) by means of a series of strong focusing magnetic quadrupole lenses (e.g. quadrupole triplet). Beam scanning takes place using magnetic or electrostatic deflection before the focusing lenses, and is driven by a feedback signal derived from the proton interactions with the resist. This feedback mechanism ensures a constant beam exposure per pixel as the beam is scanned across the resist, resulting in high-quality structures [89].

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The technology, which uses a MeV proton beam focused to small spot sizes and scanned across a resist, is similar to that developed for nuclear microscopy and nuclear microprobe applications using MeV protons¹⁴. A small particle accelerator provides a stable beam of MeV ions that is focused using a system of strong focusing quadrupole lenses. In p-beam writing, the beam is scanned across a resist material in a predetermined pattern, which is subsequently developed to produce three-dimensional structures. The CIBA setup is shown schematically in Figure 2-2 [89]. The fabrication process will vary according to the size and complexity of pattern. In our case, several hours are required.

The CGH was patterned in an effective area of $2\text{ mm} \times 2\text{ mm}$ containing 1024×1024 pixels ($2\text{ }\mu\text{m}$ resolution). Figure 2-3 (c) shows the morphologies of the CGH on PMMA film observed from an optical microscope with low ($10\times$) magnifications. The direct comparison between the simulated and PMMA pattern was shown in Figure 2-3(d). As the image of PMMA pattern was taken from the inverse view, those two images were symmetric to the axis of symmetry marked in the figure. The same zones in simulated and PMMA pattern were marked by red circles for easier identification. The black and white colors in simulated pattern correspond to the PMMA and non-PMMA regions in PMMA pattern, respectively.

Figure 2-3 Phase distributions of the CGH with “NTU” encoded, and (b) simulated reconstructed image of the CGH, and optical microscopic images of PMMA with CGH phase pattern encoded by proton beam with (c) low ($10\times$) magnifications. (d) The direct comparison between the simulated and PMMA pattern.

The ITO glass with proton-beam patterned PMMA was then used to assemble a LC cell with a normal ITO glass. The inner surface of this counter ITO glass substrate without PMMA was coated with a polyimide (PI) layer and rubbed. Then the LC cell was filled with the E7 liquid crystal (from Merck, with an ordinary refractive index of $n_o=1.5216$ and birefringence of $\Delta n=0.2248$). The liquid crystal molecules are aligned in the rubbing direction and cell gap d is $18\text{ }\mu\text{m}$.

Figure 2-4 shows the schematic cross section of the LC CGH sample. For normal incident light, the relative phase difference ($\Delta\delta$) between the region with PMMA and region without PMMA can be written as:

$$\Delta\delta = \frac{2\pi}{\lambda} [(n_{eff1}(d+h) - (n_{eff2} \cdot d + n_p \cdot h)], \quad (2.7)$$

where d is the cell gap ($18\text{ }\mu\text{m}$), h is the thickness of PMMA film ($5\text{ }\mu\text{m}$), λ is the wavelength, $n_p=1.49$ is the PMMA refractive index, n_{eff1} and n_{eff2} are the effective LC refractive indices in the region without and with PMMA, respectively.

Figure 2-4 Schematic cross section of the LC cell with one PMMA pattern glass.

The detail experiment setup of image reconstruction from the LC cell CGH sample is shown in Figure 2-5. A He-Ne laser with wavelength of 633 nm was used as the light source. A polarization was used to control the polarization of incident laser light to be parallel to the rubbing direction of the cell. A pair of object lens and a pin hole was functioned as a spatial filter and a lens behind the spatial filter acted as a collimator. The CGH sample was place before the Fourier transform lens and its image after the Fourier transform lens was enlarged by an objective lens and finally projected in the image plane.

Figure 2-5 Optical experiment setup for image reconstruction from the LC cell CGH sample.

It is necessary to point out that, because the PMMA substrate is not rubbed, the actually LC orientation on this substrate, where the anchoring is

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more complicated in contrast to the other substrate with rubbing, is not uniform and could be affected by the PMMA walls. This non-uniform orientation of LC was observed by checking the CGH sample under an optical microscope through two cross polarizers with rubbing direction of the cell parallel to the optical axis of one polarizer. Moreover the sharp edges of PMMA CGH pattern could result in local dislocations or defects. Due to the irregular pattern of the CGH, at present, it is not possible to consider all these factors in our simulation to determine the phase change as a function of voltage. Thus, in the following model, we assume a homogenous alignment assumption (anti-parallel rubbing). Though there is some discrepancy between the simulation and experiment, our model is still able to match the experiment well to certain extent.

2.2.2 Simulation Model of LC cell with PMMA substrate

As described, when the applied voltage is added on the LC cell, the LC molecules are forced to align along the direction of the electric field in the cell. Following the Oseen-Frank theory, the elastic energy density of a deformed LC can be written by [90]:

$$U_{EL} = \frac{1}{2} K_{11} (\nabla \cdot \mathbf{n})^2 + \frac{1}{2} K_{22} (\mathbf{n} \cdot \nabla \times \mathbf{n})^2 + \frac{1}{2} K_{33} (\mathbf{n} \times \nabla \times \mathbf{n})^2, \quad (2.8)$$

where $\mathbf{n}$ is a unit vector representing the LC molecule director in the cell, K_{11} , K_{22} , K_{33} are the splay, twist, and bend elastic constants, respectively.

The electromagnetic energy density can be expressed as [90]:

$$U_{EM} = \frac{1}{2} \mathbf{E} \cdot \mathbf{D} \quad (2.9)$$

where $\mathbf{E}$ is electric field vector and $\mathbf{D}$ is the displacement field vector.

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We chose homogeneous alignment configuration to numerical model the LC electro-optics effect, and the molecule director is [90]

$$\mathbf{n} = (\cos \theta, \theta, \sin \theta), \quad (2.10)$$

where θ is the orientation angle between the director and the x axis (shown in Figure 2-4). Under the homogeneous alignment condition, the elastic and the change of electromagnetic energy density can be written as [90]:

$$U_{EL} = \frac{1}{2} (K_{11} \cos^2 \theta + K_{33} \sin^2 \theta) \left(\frac{d\theta}{dz} \right)^2 \quad (2.11)$$

$$\Delta U_{EM} = \frac{1}{2\epsilon_0} \frac{D_z^2}{(\epsilon_{\parallel} \sin^2 \theta + \epsilon_{\perp} \cos^2 \theta)} - \frac{1}{2\epsilon_0} \frac{D_z^2}{\epsilon_{\perp}} \quad (2.12)$$

The total free energy including elastic energy and electromagnetic energy in the cell can be given by [90]:

$$U = \int_0^L \left[\frac{1}{2} (K_{11} \cos^2 \theta + K_{33} \sin^2 \theta) \left(\frac{d\theta}{dz} \right)^2 + \frac{1}{2\epsilon_0} \frac{D_z^2}{(\epsilon_{\parallel} \sin^2 \theta + \epsilon_{\perp} \cos^2 \theta)} - \frac{1}{2\epsilon_0} \frac{D_z^2}{\epsilon_{\perp}} \right] dz, \quad (2.13)$$

where L is the thickness of LC cell in z direction, D_z (constant here) is the z component of the electric displacement field vector, ϵ_0 is the dielectric constant in free space, and $\epsilon_{\parallel}$ and $\epsilon_{\perp}$ are the dielectric constants of parallel and perpendicular to the molecule director of nematic LC, respectively. The LC molecule director distribution will be in the equilibrium state where the total free energy is minimized. Using techniques of calculus of variation, we obtain [90]:

$$\begin{aligned} & (K_{11} \cos^2 \theta + K_{33} \sin^2 \theta) \left(\frac{d\theta}{dz} \right)^2 \\ &= \frac{D_z^2}{\epsilon_0 (\epsilon_{\parallel} \sin^2 \theta + \epsilon_{\perp} \cos^2 \theta)} - \frac{D_z^2}{\epsilon_0 (\epsilon_{\parallel} \sin^2 \theta_m + \epsilon_{\perp} \cos^2 \theta_m)}, \end{aligned} \quad (2.14)$$

where θ_m is the maximum orientation angle at the center of the cell $z = L/2$.

Here we assumed strong anchoring at both surfaces, which means the directors at the boundaries remain parallel to the rubbed direction ($\theta(0) = \theta(L) = 0$).

Defining new variable parameters as follows [90]:

$$\begin{aligned} \sin \theta &= \sin \theta_m \sin \psi \\ \epsilon_{\parallel} \sin^2 \theta + \epsilon_{\perp} \cos^2 \theta &= \epsilon_{\perp} + (\epsilon_{\parallel} - \epsilon_{\perp}) \sin^2 \theta = \epsilon_{\perp} (1 + \gamma \eta^2 \sin^2 \psi) \\ K_{11} \cos^2 \theta + K_{33} \sin^2 \theta &= K_{11} + (K_{33} - K_{11}) \sin^2 \theta = K_{11} (1 + \kappa \eta^2 \sin^2 \psi) \\ d\theta &= \frac{\sqrt{\sin^2 \theta_m - \sin^2 \theta}}{\sqrt{1 - \eta^2 \sin^2 \psi}} d\psi \\ \eta &= \sin \theta_m \\ \gamma &= \frac{\epsilon_{\parallel} - \epsilon_{\perp}}{\epsilon_{\perp}} \\ \kappa &= \frac{K_{33} - K_{11}}{K_{11}} \end{aligned} \quad (2.15)$$

The molecule director distribution inside the cell θ_m can be calculated at a given applied voltage by numerically solving Eq. (2.16) [90]:

$$\int_0^{\frac{\pi}{2}} \frac{\sqrt{(1 + \kappa \eta^2 \sin^2 \psi)(1 + \gamma \eta^2)}}{\sqrt{(1 - \eta^2 \sin^2 \psi)(1 + \gamma \eta^2 \sin^2 \psi)}} d\psi = \frac{\pi}{2} \frac{V}{V_{cl}} \quad (2.16)$$

where [90]

$$V_{cl} = \pi \frac{\sqrt{K_{11}}}{\sqrt{\epsilon_0 (\epsilon_{\parallel} - \epsilon_{\perp})}} \quad (2.17)$$

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For a fixed θ_m , the relationship of molecule director angle θ and z can be given by [90]:

$$\frac{2z}{d} = \frac{\int_0^{\psi(z)} \left(\sqrt{(1 + \kappa\eta^2 \sin^2 \psi)(1 + \gamma\eta^2 \sin^2 \psi)} / \sqrt{1 - \eta^2 \sin^2 \psi} \right) d\psi}{\int_0^{\pi/2} \left(\sqrt{(1 + \kappa\eta^2 \sin^2 \psi)(1 + \gamma\eta^2 \sin^2 \psi)} / \sqrt{1 - \eta^2 \sin^2 \psi} \right) d\psi} \quad (2.18)$$

The molecule director distribution inside the cell θ_z can be calculated at a given applied voltage by numerically solving Eq. (2.16) and (2.18).

When the polarization direction of incident light is parallel to the rubbed direction, light in LC cell will experience extraordinary refractive index. Correspondingly, the distribution of extraordinary refractive index in LC cell and the effective refractive index can be simplified by a one-dimensional solution [90]:

$$n_{eff} = \frac{1}{L} \int_0^L \frac{n_o n_e}{(n_o^2 \cos^2 \theta_z + n_e^2 \sin^2 \theta_z)^{1/2}} dz, \quad (2.19)$$

where n_o and n_e are ordinary and extraordinary refractive index of LC, respectively.

The LC we used was E7 from Merck with $n_o = 1.5216$ and $n_e = 1.7462$. The dielectric constants of E7 are $\epsilon_{||} = 19.0$ and $\epsilon_{\perp} = 5.2$, at 1 kHz frequency and room temperature. The splay and bend elastic constants of E7 chosen here are $K_{11} = 11.01 \times 10^{-12}$ N and $K_{33} = 17.01 \times 10^{-12}$ N, respectively. Figure 2-6 (a) shows the simulated relationship between the effective refractive index and the applied voltage. The calculated theoretical threshold voltage is about $0.94 V_{rms}$.

Figure 2-6 (a) The relationship between effective refractive index and applied voltage for homogeneous alignment configuration. (b) The relationship of $V_{\text{eff}}/V_{\text{apply}}$ and ϵ_{LC} with $h=5\text{ }\mu\text{m}$ and $d=16.5\text{ }\mu\text{m}$.

When a voltage is applied on the region with PMMA of our device, the effective voltage on the LC will be reduced because PMMA is an insulator (with a dielectric constant of approximately $\epsilon_p = 3$ [91]). The effective voltage in the region with PMMA can be determined by:

$$V_{\text{eff}} = \frac{1}{1 + \frac{\epsilon_{\text{LC}} \cdot h}{\epsilon_p \cdot d}} V_{\text{apply}}, \quad (2.20)$$

where V_{apply} is the external applied voltage on the two ITO layers and ϵ_{LC} is the effective dielectric constant of LC at the interface of PMMA and LC in the region with PMMA. The relationship of $V_{\text{eff}} / V_{\text{apply}}$ and ϵ_{LC} is shown as Figure 2-6(b), from which we can see that the ratio of V_{eff} to V_{apply} decreases with the increase of ϵ_{LC} .

2.3 Electro-Optical Characteristics

As known from the Eq. (2.20), if we obtain the value of V_{apply} , we can determine the effective voltage of V_{eff} , effective refractive index of $n_{\text{eff}1}$ and $n_{\text{eff}2}$. Hence, the corresponding phase difference can be determined using Eq. (4.1). For $\epsilon_{LC} = \epsilon_{\perp} = 5.2$, we calculated that around 67.5% of the external applied voltage is finally applied on the LC for $h=5 \mu\text{m}$ and $d=16.5 \mu\text{m}$.

Figure 2-7 The solid blue line shows the theoretical relationship of phase difference and applied voltage, and red stars represent the experimental phase difference at applied voltage of $1.2 V_{\text{rms}}$, $1.32 V_{\text{rms}}$, $1.54 V_{\text{rms}}$, $1.97 V_{\text{rms}}$, $2.53 V_{\text{rms}}$, $3.18 V_{\text{rms}}$ and $7.0 V_{\text{rms}}$ respectively.

Under applied voltage, the reconstructed image would become blurred if the phase difference is the even integer times of π and the reconstructed image would become clear if the phase difference is the odd integer times of π . In our experiments, the blurred images were found at voltage of $1.2 V_{\text{rms}}$, $1.54 V_{\text{rms}}$, $2.53 V_{\text{rms}}$ and $7.0 V_{\text{rms}}$, corresponding to 0π , -2π , -2π and 0π , respectively. The clear images were observed at voltage of $1.32 V_{\text{rms}}$, $1.97 V_{\text{rms}}$, and $3.18 V_{\text{rms}}$, corresponding to $-\pi$, -3π and $-\pi$, respectively. In Figure 2-7, the solid blue line shows the theoretical relationship of phase difference and applied voltage, and red stars represent the experimental phase difference at applied voltage of $1.2 V_{\text{rms}}$, $1.32 V_{\text{rms}}$, $1.54 V_{\text{rms}}$, $1.97 V_{\text{rms}}$, $2.53 V_{\text{rms}}$, $3.18 V_{\text{rms}}$ and $7.0 V_{\text{rms}}$ respectively. We can see that the experimental data are consistent with the calculated results. In our experiment, the image started to change from $1.2 V_{\text{rms}}$, i.e. the experimental threshold was about $1.2 V_{\text{rms}}$.

Figure 2-8 Reconstructed images of the LC cell CGH sample with an applied voltage of (a) $1.2 V_{\text{rms}}$, (b) $1.32 V_{\text{rms}}$, (c) $1.54 V_{\text{rms}}$, (d) $1.97 V_{\text{rms}}$, (e) $2.53 V_{\text{rms}}$, (f) $3.18 V_{\text{rms}}$, (g) $7.0 V_{\text{rms}}$, and (h) $12 V_{\text{rms}}$ respectively.

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Figure 2-8 (a) - (h) show the reconstructed images of the LC cell CGH sample at voltages of $1.2 V_{\text{rms}}$, $1.32 V_{\text{rms}}$, $1.54 V_{\text{rms}}$, $1.97 V_{\text{rms}}$, $2.53 V_{\text{rms}}$, $3.18 V_{\text{rms}}$, $7.0 V_{\text{rms}}$ and $12 V_{\text{rms}}$ respectively. In order to reduce the scattered light from the brightest 0th-order beam in the reconstructed image, a hole was drilled in the middle of the image screen. As the applied voltage exceeds the threshold ($1.2 V_{\text{rms}}$), the image changes between clear and blurred alternately several times as shown in Figure 2-8. The blurred image is primarily due to phase mismatch, originated from non-uniform anchoring on PMMA walls and sharp edges, leading to poor/non-uniform modulation. Also the small PMMA wall spacing could generate strong anchoring LC forces leading to poor modulation capability. It is noticed that the image at higher voltage (e.g. $12 V_{\text{rms}}$) is clearer than those at lower voltages (e.g. $1.32 V_{\text{rms}}$, $1.97 V_{\text{rms}}$ and $3.18 V_{\text{rms}}$). This poor quality at lower voltage is probably resulted from a poor phase modulation and error due to complex LC orientation related to CGH pattern. In contrast, at a higher voltage, the LC directors will reorient mostly along the applied electric field and become more ordered, resulting in reduced light scattering and hence a clearer reconstructed image. For higher applied voltages ($> 12 V_{\text{rms}}$), there is no further change in the reconstructed image, indicating that most of the LC molecules, which are not anchored by PI surface, are perpendicular to the substrate. The calculated phase difference stays at around 0.57π for voltages higher than $12 V_{\text{rms}}$. The diffraction efficiency is defined by the ratio of the light intensity of the first order diffraction to the total transmitted light intensity through the CGH in the reconstructed image plane, which can be measured directly. The maximum diffraction efficiency measured

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was about 28.7% at $12 V_{\text{rms}}$, less than the theoretical maximum binary phase first order diffraction efficiency of 41% [92]. We had checked the CGH sample under an optical microscope through cross polarizers with no voltage applied. The direction of LC molecules in the region without PMMA were not exactly along the rubbing direction, which will reduce the initial phase difference between the regions with and without PMMA, leading to n_{eff} differences between theoretical and experimental n_{eff} values under applied voltages.

Although the polarization of the incident light chosen in our experiment was parallel to the rubbing direction of LC cell, i.e. an effective extraordinary refractive index of LC was experienced by the incident light, the reconstructed image of CGH can also be obtained by using incident light with polarization perpendicular to the rubbing direction of LC cell, since the ordinary refractive index of LC ($n_o = 1.521$) and the refraction index of PMMA ($n_p = 1.49$) are not equal. In this configuration, the relative phase difference $\Delta\delta$ was calculated to be 0.57π . With an applied external voltage, the relative phase difference and the reconstructed image of the CGH kept unchanged.

Figure 2-9 shows the electro-optical response of the LC cell CGH sample measured by switching the applied voltage between $4.5 V_{\text{rms}}$ and $24 V_{\text{rms}}$ with a frequency of 2.6 Hz. The rising time and falling time are about 62 ms and 60 ms respectively. Reducing the LC cell gap would lead to a faster response time.

Figure 2-9 Electro-optical response of the CGH encoded LC cell.

2.4 Conclusion

In conclusion, we demonstrated an electrically-switchable CGH using LC cell with proton beam patterned PMMA film. The CGH desired for generating words of NTU was firstly generated by computer and then fabricated on PMMA substrate coating on ITO glass by proton beam writing method. The advantages of proton beam writing (PBW) used here are that it has a deep penetration in resist materials with little lateral scattering which enables PBW to fabricate high aspect ratio microstructures with vertical, smooth sidewalls and low line edge roughness. The highest resolution achieved is around 50 nm and a 2-MeV proton will penetrate about 60 μm into PMMA. The high aspect ratio could be used to fabricate Bragg diffraction volume grating with high diffracting efficiency. Comparing to the CGH recorded in PDLC where a transparency mask was used with a resolution of 50 μm [84], a higher resolution ($\sim 2\mu\text{m}$) was achieved by proton beam, leading to a better quality of

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the reconstructed CGH image. It has less light scattering and relatively lower operating voltage ($\sim 15 V_{\text{rms}}$), compared to the CGH using a PDLC material [84]. Higher cost and longer fabricate time are the drawbacks of this method. The maximum diffraction efficiency measured is about 28.7%. This device is promising in adaptive optical elements.

Chapter 3 Liquid Crystals Based Tunable Finite Energy Airy Beams Generator

3.1 Introduction

3.1.1 Airy Function

Airy functions are special functions, which is named after the British astronomer George Biddle Airy. In physics, the Airy function is the solution to Schrödinger's equation for a particle confined within a triangular potential well and for a particle in the one-dimensional constant force field. The function of $Ai(x)$ and the relative function of $Bi(x)$ are the solutions of a differential equation as following:

$$\frac{d^2 y}{dx^2} - xy = 0. \quad (3.1)$$

The expression of $Ai(x)$ is one solution of Eq. (3.1) and defined by:

$$Ai(x) = \frac{1}{\pi} \int_0^\infty \cos\left(\frac{1}{3}t^3 + xt\right) dt \quad (3.2)$$

Another solution of Eq. (3.1) is denoted as $Bi(x)$ and is given by:

$$Bi(x) = \frac{1}{\pi} \int_0^\infty \left[\exp\left(-\frac{1}{3}t^3 + xt\right) + \sin\left(\frac{1}{3}t^3 + xt\right) \right] dt. \quad (3.3)$$

Figure 3-1 shows the plot of functions $Ai(x)$ and $Bi(x)$.

Figure 3-1 Plot of $Ai(x)$ and $Bi(x)$ functions.

3.1.2 Airy Wave Packet

In 1979, Berry and Balazs firstly reported that the force-free one-dimensional Schrödinger's equation [93]:

$$i\hbar\partial_t\psi(x,t) + \frac{\hbar^2}{2m}\partial_x^2\psi(x,t) = 0, \quad (3.4)$$

has a non-spreading unique packet solution with expression of Airy function as [93]:

$$\psi(x,t) = Ai\left[\frac{B}{\hbar^{2/3}}\left(x - \frac{B^3 t^2}{4m^2}\right)\right] e^{(iB^3 t/2m\hbar)[x - (B^3 t^2/6m^2)]}. \quad (3.5)$$

where B is an arbitrary constant, $\hbar$ is Dirac constant. At $t=0$, we have the wave packet [93]

$$\psi(x,0) = Ai\left(\frac{Bx}{\hbar^{2/3}}\right). \quad (3.6)$$

The square of the envelope $|\psi(x,t)|^2$ not only keeps unchanged in form but also continually accelerates even without force acts.

The acceleration process associated with these Airy wave packets was later investigated by Greenberger [94], and such investigation work was

extended to 3D Airy wave packet situation by I. M. Besieris etc [95]. Although the useful and unique property of Airy wave packet, the Airy wave's oscillating tail decays very slowly even the input of x is approaching infinite because of the nature of Airy function, it is very difficult to synthesize such wave function unless its amplitude has been truncated.

3.1.3 Finite Energy Airy Beams

A breakthrough of utilizing Airy wave packet was achieved by G. A. Siviloglou, and D. N. Christodoulides [96] in 2007, who released a paper discussing about how to realize Airy beam by adding an exponentially decaying item on the expression of Airy function as well as the propagation property of such finite energy Airy beam in linear condition. In their paper, they investigated the acceleration dynamics of quasi-diffraction-free finite energy Airy beams, which can retain their intensity features over several diffraction lengths and still can accelerate in the transverse direction [96].

The analysis starts from so called (1+1) D paraxial equation of diffraction, which governs the propagation dynamics of electric field envelope $\phi(s, \xi)$ related to planar optical beams [96]:

$$i \frac{\partial \phi}{\partial \xi} + \frac{1}{2} \frac{\partial^2 \phi}{\partial s^2} = 0, \quad (3.7)$$

where $s = x/x_0$ is a dimensionless transverse coordinate, x_0 represents an arbitrary transverse scale, $\xi = z/kx_0^2$ represents a normalized propagation distance, $k = 2\pi n/\lambda_0$ represents the wave number of optical wave, n is the refractive index of medium optical wave travels within, and λ_0 is the wavelength of optical wave in vacuum.

The exponentially decaying version of Airy beams at the input of system is defined as:

$$\phi(s, \xi = 0) = Ai(s) \exp(as), \quad (3.8)$$

where a is a positive quantity to ensure containment of the infinite Airy tail and thus makes true of physical realization of such beams. With that exponent item adding on Airy function, the positive branch of Airy beam intensity decays rapidly, thus leading to a guarantee of the convergence of electric field envelope function in Eq. (3.8). An example of normalized field profile and intensity profile of finite Airy beam at $\xi = 0$ ($z=0$) is shown in Figure 3-2 [96].

Figure 3-2 (a) Normalized field profile, and (b) normalized intensity field profile of a finite energy Airy beam with $a=0.1$ [96].

By directly solving Eq. (3.7) with the initial conditions of Eq. (3.8), the Quasi Airy Beam will evolve according to:

$$\phi(s, \xi) = Ai[s - (\xi/2)^2 + ia\xi] \exp[as - (a\xi^2/2) - i(\xi^3/12) + i(a^2\xi/2) + i(s\xi/2)]. \quad (3.9)$$

In most of cases, the value of positive $a \ll 1$ is very small, thus leads to a similarity of intensity profile between produced finite energy Airy beam and original Airy beams. Figure 3-3 shows the propagation dynamics of a diffractive-free original Airy wave and corresponding finite energy Airy beam

with $a=0.05$, where both intensities of beams are shown [97]. In an extreme case, at the limit of $a=0$, the solution in Eq. (3.9) reduces to the non-dispersive and diffraction-free Airy wave packet.

Figure 3-3 Propagation dynamics of (a) a diffraction-free Airy wave, and (b) a finite energy Airy beam with $a=0.05$. The corresponding input intensities are shown in the insets [97].

The Fourier spectrum of this finite energy Airy beam at input of system, Eq. (3.8), is of in our interest which can be used to generate such beams through Fourier transform and can be expressed by:

$$\Phi_0(k) = \exp(-ak^2) \exp\left(\frac{i}{3}(k^3 - 3a^2k - ia^3)\right). \quad (3.10)$$

Eq. (3.10) indicates that the wave-packet power spectrum is Gaussian. When a is small, the second and third items in exponential phase expression can be ignored and thus the Fourier transform $\Phi_0(k)$ of this finite energy Airy wave packet can be given as

$$\Phi_0(k) = \exp(-ak^2) \exp\left(\frac{i}{3}k^3\right), \quad (3.11)$$

which means that the Fourier spectrum of truncated Airy beam can be expressed by combination of Gaussian with a cubic phase distribution. It is important implications of experimentally synthesizing finite energy version of Airy packet. As a result, the finite energy Airy beams can be created from a broad Gaussian beam imposed a cubic phase through a Fourier transformation [97]. From Parseval's theorem, the total power of this finite energy Airy wave packet can be obtained from [96]:

$$\int_{-\infty}^{\infty} ds |\phi(s, \xi = 0)|^2 = \sqrt{\frac{1}{8\pi a}} \exp\left(\frac{2a^3}{3}\right). \quad (3.12)$$

For two-dimensional case, the expression of electric field envelope can be easily deduced from the 1D case, as:

$$\phi(s_x, s_y, \xi = 0) = Ai(s_x) Ai(s_y) \exp[a(s_x + s_y)], \quad (3.13)$$

where $s_x = x / x_0$, and $s_y = y / y_0$.

An example of intensity profiles of 2D finite energy Airy beam at different propagation position are shown in Figure 3-4[96]. In contrast of

propagation along one direction, the 2D beam propagates along a 45° between x and y axis.

Figure 3-4 Intensity profile of 2D finite energy Airy beam at (a) $z=0$ and (b) $z=50$ cm. Where $x_0=y_0=100$ μm , and $a=0.1$ [96].

The increasing concern on Airy beams of scientists led to a lot of research reports and investigations both in theory and experiment recently [98-101]. Unique properties of Airy beams include ballistic dynamics in free space [102], self-healing [103], Poynting vector and angular momentum [104]. Studies also extended to investigate skewed, nonspreading, accelerating Airy wave packets with obliquity factor in bidispersive optical media [105], new types of finite energy Airy beams with versions of Ai and Bi Airy functions [106], propagation dynamics of an optical pulse imposed on an Airy beam [107], new form of Airy beam composed of two symmetrical Airy beams [108], and Kerr nonlinearity effect on Airy beam [109].

3.1.4 Realization and Applications

As mentioned above, the Airy beam can be generated from the Fourier transform of a Gaussian function with a phase modulation item, which gives the realization of such beams a lot of freedoms. The summary of different kinds of

realization methods is following: In 2008, P. Saari [110] studied the properties of laterally accelerating Airy generated in forms of pulses. In 2009, T. Ellenbogen etc [111] reported an Airy beam generated by three-wave mixing processes, from an asymmetric structure quadratic nonlinear optical media. In 2010, more reports were released on the realization methods, I. Dolev eat [112, 113] investigated the realization and switching of Airy beam in a quadratic nonlinear photonic crystals also using three wave mixing technique. S. Jia etc [114] realized diffusion-trapped Airy beams in photorefractive media by two-wave mixing technique. D. G. Papazoglou etc [115] demonstrated the method of generating both one- and two-dimensional tunable Airy beams using adjusted cylindrical lenses. A. Salandrino etc [116] introduced the Airy beam in a new form of plasmon as a nondiffracting surface wave with potential applications in plasmonic energy routing. Y. Hu etc [117] studied the persistence and breakdown of Airy beams driven by an initial nonlinearity when propagating from a nonlinear medium to a linear medium. Y. Gu etc [118] investigated the scintillation of Airy beam arrays in atmospheric turbulence by utilizing the self-bending propagation property. D. Abdollahpour etc [119] reported the realization of intense Airy-Airy-Airy light bullets by combing a spatial Airy beam with an Airy pulse in time. And in 2011, S. Longhi [120] generated Airy beams using a microchip laser with plane-plane emit.

Figure 3-5 Micro photographs of particles exposure in Airy beam. The incident direction of Airy beam is from up-right side to left bottom side [121].

After theoretically and experimentally works on unique properties of Airy beams, the application potential of such beam with its characteristics comes in front of people. One possible application of Airy beams was firstly found by J. Baumgartl, M. Mazilu, and K. Dholakia [121], who exploited a new method in optical micromanipulation by using Airy beams. Micro particles and cells can be cleared from a sample chamber as well as precisely driven in a parabolic trajectory due to the Airy beams. Figure 3-5 shows the image of particles driving by Airy beams. Beside application in optical trapping, another possible application is about the generation of curved plasma channel using ultraintense Airy beams, which was reported by P. Polynkin etc [122] in 2009. The experiment setup for curved plasma channels from femtosecond Airy beams was shown in Figure 3-6. Other applications of Airy beams reports include using Airy beams as versatile linear light bullets [123], optical trapping [124], electron capture [125], vacuum electron acceleration driven [126], and vacuum laser driven acceleration [127].

Figure 3- 6 Experiment setup for curved plasma channels from femtosecond Airy beams [122].

3.2 Fabrication of Liquid Crystal Cell with Cubic Phase

Several media including spatial light modulator (SLM) [97], continuous phase mask [122], and even nonlinear photonic crystal were reported to generate Airy beams previously [112, 113]. However, all of these methods mentioned above possessed either high cost or complicated fabrication process. In contrast, optical elements with binary-phase pattern could be fabricated easily with lower cost, which provides a simple way to generate optical beams such as Airy beams. Therefore, it is useful and meaningful to study the properties of Airy beams produced by such optical binary-phase elements.

On the other hand, liquid crystal (LC) has tunable refractive properties, and it has been applied in realizing tunable diffractive optical elements (e. g., Fresnel zone plate [128- 131], optical vortex [132, 133], etc.). In our group, the polymer-dispersed liquid crystal (PDLC) based binary-phase pattern was used

Chapter 3 Liquid Crystals Based Tunable Finite Energy Airy Beams Generator

to generate Airy beam previously [134], however, the phase modulation is only about 0.3π , and a larger phase modulation cannot be achieved using PDLC. In this section, we demonstrate electrically switchable finite energy Airy beams generated by a liquid crystal cell with binary-phase patterned electrode. Both of the electrical tunable properties and the propagation characteristics of the Airy beams are investigated in detail.

In 2D case, the initial field envelope of finite-energy Airy wave packets can be given by: $\phi(s_x, s_y, \xi = 0) = Ai(s_x)Ai(s_y)\exp[a(s_x + s_y)]$, where $s_x = x/x_0, s_y = y/y_0$. The Fourier transform can be written as: $\Phi_0(k_x, k_y) \propto \exp[-a(k_x^2 + k_y^2)]\exp[i(k_x^3 + k_y^3)/3]$, where k_x and k_y are Fourier spectrum coordinates.

Figure 3-7(a) shows a 2D continuous phase pattern of an Airy wave packet. The 2D continuous phase pattern for 2D Airy beam is generated from the phase item $(k_x^3 + k_y^3)/3$, while both k_x and k_y ranges from $-\sqrt[3]{17.25\pi}$ to $+\sqrt[3]{17.25\pi}$. Then the generated 2D phase distribution has been wrapped between 0 and 2π , resulting a pattern varies from -11.5π to $+11.5\pi$ along diagonal in Figure 3-7 (a) with size of 0.43 cm. In our experiment, the binary-phase pattern with cubic phase modulation was obtained by converting the continuous phase pattern by setting the value of phase delay between $0 \sim \pi$ as 0 and $\pi \sim 2\pi$ as π , respectively. This binary-phase pattern is shown in Figure 3-7(b), where the black and white regions represent phase values of 0 and π , respectively. Then, the binary-phase pattern was transferred to a transparent mask, using which, a patterned indium-tin-oxide (ITO)-coated glass substrate was obtained by a standard photolithography process.

The photolithography process includes 5 steps: photoresist coating, exposure, developing, etching, and stripping. There are two types of photoresist: positive and negative. For positive resists (used in our experiment), the resist is then exposed covered by photomask with UV light wherever the exposed part is to be removed; for negative resists, the exposed resist is to be left. The exposed resist is then washed away by the developer solution, leaving unexposed resist on ITO glass. In etching processes, the resist film that remains after the development functions as a protective mask on ITO glass while the ITO in bared areas is being etched. The remaining resist film is finally stripped, leaving a pattern of the desired ITO pattern in the glass substrate.

The patterned ITO glass was then used to assemble a LC cell with a blank ITO glass as the counter electrode. The LC cell was prepared by anti-parallel rubbing. The cell gap was $d = 7.5 \mu\text{m}$ controlled by spacers. The liquid crystal used in our experiment was E7 from Merck, with $n_o = 1.5216$ and $n_e = 1.7462$.

Figure 3- 7 (a) Continuous phase mask and (b) the corresponding binary-phase mask used to generate the 2D Airy beam. Optical microscopic images of the LC cell sandwiched between two cross polarizers at applied voltage of (c) $3 V_{rms}$ and (d) $9 V_{rms}$. The Scale bar in (c) and (d): 0.5 cm.

When the voltage is applied on the LC cell, LC molecules in the region with ITO electrode will reorient according to the external voltage, while the LC

molecules in the region without ITO electrode will remain unchanged. The schematic of LC cell with binary phase structure was shown in Figure 3-8, under the condition of (a) without and (b) with external applied voltage.

Figure 3-8 Schematic of LC cell with binary phase pattern while (a) without voltage, and (b) with voltage applied.

Figure 3-7(c) and (d) show the optical microscopic images of the LC cell sandwiched between two cross polarizers at applied voltage of $3 V_{rms}$ and $9 V_{rms}$, respectively. The rubbing direction of LC cell was at 45^0 with respect to both polarizer and analyzer axes. As the voltage exceeds the threshold ($\sim 0.81 V_{rms}$), the LC directors in the regions with ITO electrode start to reorient, as reflected by the color change as shown in Figure 3-7(c). In the high voltage regime ($\sim 9 V_{rms}$), the LC directors in the regions with ITO electrode are reoriented nearly perpendicular to the substrates, leading to a black state as shown in Figure 3-7(d).

3.3 Optical Setup For Generating Airy Beams

If the polarization of the incident light is parallel to the rubbing direction of the cell, the phase difference ($\Delta\delta$) between the regions with and without ITO electrode under the external voltage can be expressed as $\Delta\delta = \frac{2\pi}{\lambda}(n_e - n_{eff}) \cdot d$, where d is the cell gap, λ is the wavelength of incident light, and n_{eff} is effective LC refractive index in the region with ITO electrode.

Figure 3-9 Optical setup for Airy beam reconstruction. BE, beam expander; L, lens.

The LC cell sample recorded with binary cubic phase pattern was illuminated by a collimated He-Ne laser (633 nm) with a beam diameter of about 1 cm to generate the finite energy Airy beams. A polarizer was used to ensure the polarization of incident light was parallel to the rubbing direction of LC cell sample. A spherical lens ($f=20$ cm) placed at a distance of f behind the LC cell sample was used to perform the Fourier transform of the binary phase pattern. The Fourier transform or finite energy Airy beam was obtained at a distance of $d=f$ behind the lens, and then captured by a charge-coupled-device (CCD) camera. The optical setup is illustrated in Figure 3-9.

3.4 Electrically Tunable Property of Liquid Crystal Based Airy Beams

The initial phase difference between the regions with and without ITO electrode is zero. The Airy beams would be generated (e.g. at voltages of $1.08V_{rms}$, $1.69V_{rms}$, and $6.0V_{rms}$) when the phase difference is the odd integer times of π , and the Airy beams would be erased (e.g. at voltages of $1.31V_{rms}$, $2.29V_{rms}$) when the phase difference is the even integer times of π . Figure 3-10(a), (b), and (c) show the observed intensity distribution of 2D Airy wave packet under voltages of $1.08V_{rms}$, $1.25V_{rms}$ and $1.31V_{rms}$, at $z = 9$ cm, respectively. We can see that, the 2D finite energy Airy beams were generated in Figure 3-10(a), blurred in Figure 3-10(b), and erased in Figure 3-10(c).

Figure 3-10 The observed intensity distribution of the 2D Airy wave packets under different voltages of (a) $1.08V_{rms}$, (b) $1.25V_{rms}$, and (c) $1.31V_{rms}$, at $z = 9$ cm.

It was worth mentioning that the generated image consisted of a pair of symmetric Airy beams with opposite propagation directions, which was due to the intrinsic property of binary element. The coordinate system is also labeled in Figure 3-10(a). When the applied voltage was higher than $6.0V_{rms}$, the generated image did not show significant change, indicating the LC directors in the region with ITO electrode were reoriented almost along the electric field and the phase difference would not change anymore.

3.5 Simulation of Accelerating Propagation of Airy Beams

To better understand the 2D finite energy Airy beams from binary-phase element, several simulated images in x-y plane, calculated using Angular Spectrum Method from GWO library [19, 20], are shown in Figure 3-11(a)-(d) at different distances of $z = 4$ cm, 7 cm, 8.5 cm, and 10 cm, respectively. The simulation was used to describe the propagation of the Airy beam generated by the binary-phase pattern, its parameters were set according to real experimental setup with binary-phase pattern sample size of $0.43\text{ cm} \times 0.43\text{ cm}$, a Fourier transform lens ($f = 20\text{ cm}$), and π phase difference between the regions with and without ITO electrode. The corresponding images obtained from experiment at applied voltage of $1.08V_{\text{rms}}$ ($\Delta\delta = \pi$ when voltage is at $1.08V_{\text{rms}}$) are also shown in Figure 3-11(e)-(h), respectively, which are consistent with the simulation images. Because of the twin lobes of Airy beams generated from binary element, the two Airy beams maximum intensity points were overlapped (Figure 3-11(e)) before $z=6.5$ cm. After that distance, these twin Airy beams then separated with each other and propagated along the bisection direction of x- and y-axis with opposite directions (Figure 3-11(f)-(h)). This overlap property of the twin Airy beams arisen from the binary-phase element is not possessed by the Airy beam produced using spatial light modulator (SLM) with continuous phase modulation. To obtain the detail transverse profile of the Airy beam, an objective lens was located just before the CCD camera for magnification purpose. Figure 3-12 shows the magnified transverse profile of the 2D finite energy Airy beam under voltage of $1.08V_{\text{rms}}$, at $z=10.5$ cm. Only one group of Airy beam was captured here.

Figure 3-11 Simulated images of Airy beams in x - y plane at distance of (a) $z = 4$ cm, (b) 7cm, (c) 8.5 cm, and (d) 10 cm, setting phase difference of π ; Corresponding experimental images at distance of (e) $z = 4$ cm, (f) 7 cm, (g) 8.5 cm, and (h) 10 cm, at applied voltage of $1.08V_{\text{rms}}$.

Figure 3-12 Magnified transverse profile of the two dimensional finite energy Airy beam captured by a CCD with an objective lens under voltage of $1.08V_{\text{rms}}$, at $z=10.5$ cm.

In order to study the propagation dynamics of the 2D finite energy Airy wave packet, we further studied the relationship between the deflection along the bisector direction of x - and y -axis and propagation distance (Figure 3-13), where the voltage applied on the LC cell was fixed at $1.08 V_{rms}$. In Figure 3-13, the red triangles represent simulated results based on beam propagation simulation. The blue circles represent the experimental results and the green line is a parabolic fit. We can see that the relationship of the deflection and propagation distance is parabolic, and the experimental results are quite consistent with the simulation. The parabolic relationship indicates that the Airy beams produced from binary-phase element possess a transverse acceleration, as the same as the Airy beams generated from continuous phase modulation.

Figure 3-13 Acceleration of the 2D Airy beam along the bisector direction of x - and y -axis as a function of distance. The red triangles represent simulated results, the circles represent the experimental results and the green line shows the parabolic fit curve.

In our experiment, the Airy beams moved on a 2D parabolic trajectory with $x_d = y_d$, where x_d and y_d were the transverse deflections in x and y

directions respectively. This parabolic trajectory is a result of acceleration and can be well described by $x_d \cong \lambda_0^2 z^2 / (16\pi^2 x_0^3)$ (the parabolic fit in Figure 3-13) [2]. The deflection in Figure 3-13, which represents the parameter of $\sqrt{2}x_d$, shows a shift of 459.5 μm within 5.5 cm propagation distance. The value of $x_0=45.2 \mu\text{m}$ was calculated here and the Airy beams propagated almost diffraction-free up to a distance of ~ 12 cm. The relatively small propagation distance should be caused by the small focal length of lens and the big size of LC sample.

3.6 Conclusion

In conclusion, a pair of electrically switchable 2D finite energy Airy beams has been generated by a liquid crystal cell with binary-phase patterned electrode. These Airy beams are electrically switchable, which can be generated or erased with the change of applied electric field. The diffraction-free and transverse acceleration dynamics of the Airy beams were verified. The experimental obtained deflections of propagation were quite consisted with simulated results. This kind of binary-phase element based Airy beam device has the advantages of low cost, easy fabrication process, and electrical tunability, potentially useful in adaptive optics.

Chapter 4 Liquid Crystals Based Polarization-

Dependent Circular Dammann Grating

4.1 Introduction

4.1.1 Introduction of Circular Dammann Grating

Dammann grating is a pure-phase modulation grating whose phase transition points are designed with optimization to generate equal-intensity spots at diffractive orders with high efficiency as a form of one-dimensional spots or two-dimensional rectangular spots. The property of Dammann grating makes it useful for spots array generators, and image duplication applications [135].

The concept of Circular Dammann grating was firstly proposed by C. Zhou etc [136] in 2003, which can be used to produce circular equal intensities at various orders in the far field. However, the circular Dammann grating reported by C. Zhou is only a diffractive optical element without periodic structures, thus not a grating in a strict sense. In 2006, a more precisely defined circular Dammann grating (CDG) was suggested by S. Zhao, and P. S. Chung [137]. As the phase modulation of a phase grating can be binary, multilevel, and continuous, in the paper of S. Zhao, the authors started their discussion from a binary phase circular grating without losing generality for the sake of clarity and simplicity. Following the pioneer works of C. Zhou and S. Zhao, CDG were realized by using binary [138, 139], and multilevel [140] phase modulation with different methods include Hankel transform method [137, 138] and circular spot rotation method [139]. As a useful optical element, CDG can be used in areas of focal-length measurement [141], collimation testing of beam

[142], angle measurement [143], optical image coding [144], area measurement at long-distance [145], and so on.

4.1.2 Design of Binary Circular Dammann Grating

As mentioned above, the circular Dammann grating is a periodic structure with phase transition within. Here, we introduce the design process of binary phase circular Dammann grating using Hankel transform method. Figure 4-1 shows the general profile with normalized phase transitions $\{r_k\}$ in a period length, with boundary values of $r_0=0$ and $r_N=1$ [137].

Figure 4-1 Transition points of a binary phase CDG in one period, where ϕ_0 represents phase level and is normally equal to π [137].

For a radially periodic image of $g(r)$, assuming period length is T , the Fourier spectrum is consisted by a series of half-order derivative impulses [137]. The radii of these concentric rings are integer multiples of $f=1/T$, meaning that the spectrum of radially periodic image can also be expressed a series of terms, with the n th impulse ring is represented by the n th term explicitly. In polar coordinates, the elementary radially periodic function is a circular sine function, whose Hankel transform (Fourier transform in polar coordinates) can be expressed by [146]:

$$\sin(2\pi fr) \xleftrightarrow{H} \frac{f}{\sqrt{\pi}} \frac{1}{(f+q)^{3/2}} \delta^{(1/2)}(q-f), \quad (4.1)$$

where $q=\rho/\lambda f$ is the spatial frequency, with ρ representing a polar coordinate in the observed plane and f representing the focal length of the lens, $\delta^{(1/2)}(x)$ is the half-order derivative of the impulse $\delta(x)$.

A radially periodic image $g(r)$ with period T can be decomposed into a linear combination of circular sinusoid functions as [137]:

$$g(r) = \sum_{n=1}^{\infty} c_n \sin(2\pi \frac{n}{T} r), \quad (4.2)$$

where the n th term represents the n th ring, and c_n are sine series coefficients with expression of following[137]:

$$c_n = \frac{2}{T} \int_T g(r) \sin(2\pi \frac{n}{T} r) dr, \quad (4.3)$$

and its Fourier spectrum can be calculated directly by [137]:

$$G(q) = \frac{1}{\sqrt{\pi}} \sum_{n=1}^{\infty} c_n \frac{n/T}{(n/T+q)^{3/2}} \delta^{(1/2)}(q-n/T), \quad (4.4)$$

which represents a series of concentric impulse rings with different amplitudes.

The radially periodic function $g(r)$ has an intrinsic property that the radial period has an antisymmetric profile respect to the midperiod points, e. g. the transitions have the relationship of $r_k+r_{N-k}=1$, where $k=1,2,\dots,N/2-1$ represents the transitions in the first half-period. The value of N is an even number and $r_{N/2}=0.5$ is a transition. Under such restrictions of CDG property, the sine series coefficients c_n can be obtained from Eq. (4.3) [137]

$$c_n = \frac{2}{n\pi} \left[2 \sum_{k=1}^{N/2-1} (-1)^k \cos(2\pi n x_k) + (-1)^{n+N/2} + 1 \right]. \quad (4.5)$$

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In the Fourier spectrum of $g(r)$, the intensity expression of n th pulse ring is given by [137]:

$$I_n = \left| \frac{1}{\sqrt{\pi}} c_n \frac{n/T}{(n/T + q)^{3/2}} \right|^2. \quad (4.6)$$

From the expression of intensity profile, we can obtain the equal-intensity rings condition of neighboring impulse rings should satisfied:

$$I_n = I_{n+1}, \quad (4.7)$$

which leads to the relationship of c_n and c_{n+1} as following [137]

$$\frac{|c_{n+1}|^2}{|c_n|^2} = \frac{n+1}{n}, \quad (4.8)$$

where $n=1,2,3,\dots$, denotes the order of impulse rings generated in the image plane. The goal in designing the CDG is transferred to solve the solutions of Eq.(4.8) through optimizing the parameters of the normalized transitions $\{r_k\}$ of grating.

For optimizing the phase transitions with small order, usually the iterative deepest-descent algorithms were used to calculate. The relative intensity uniformities and diffraction efficiencies are monitored by the cost function in the numerical simulation with expression of [137]:

$$\Phi = \sum_{i=1}^N (I_i - I_{av})^2 / \sum_{i=1}^N I_i, \quad (4.9)$$

where the intensities are only summed from the uniform-intensity order, and I_{av} is the average intensity of those rings. We can also define the efficiency and uniformity as following [137]:

$$\eta = \frac{1}{2} \sum_{n=1}^N |c_n|^2, \quad (4.10)$$

$$uni = \frac{\max(I_n) - \min(I_n)}{\max(I_n) + \min(I_n)}. \quad (4.11)$$

If the radially periodic function of $g(r)$ is decomposed into a set of circular cosine functions, the coefficients is given by [137]:

$$c_n = \frac{2}{T} \int_T g(r) \cos(2\pi \frac{n}{T} r) dr, \quad (4.12)$$

thus its spectrum can be given by [137]

$$G(q) = \frac{c_0}{2} \frac{1}{\pi |q|} \delta(q) + \frac{1}{\sqrt{\pi}} \sum_{n=1}^{\infty} c_n \frac{n/T}{(n/T + q)^{3/2}} \delta^{(1/2)}(n/T - q), \quad (4.13)$$

In this situation, the transition points now are symmetric with respect to midperiod transition point, transitions have the relationship of $r_k + r_{N-k} = 1$, where $k=1, 2, \dots, (N-1)/2$ represents the transitions in the first half-period. Contrast to the case of sine function, here the number of transition N should be odd. The cosine coefficients are given by [137]:

$$c_0 = 2 \left[(-1)^{N/2} - 4 \sum_{k=1}^{N/2} (-1)^k x_k \right], \quad (4.14)$$

$$c_n = \frac{-4}{in\pi} \sum_{k=1}^{N/2-1} (-1)^k \sin(2\pi n x_k). \quad (4.15)$$

4.1.3 Azo Dye Doped Liquid Crystals

Azo-dye is a kind of photosensitive material, which is capable of aligning LC molecules through a photoisomerization molecular reorientation process [147]. Azo-dye molecules reorient depending on the local polarization of light passing through by the effect of photoactivated isomerisation [148]. The realignment of the azo-dye occurs during light exposure within its absorption spectrum since its anisotropic photoisomeric states. The excess absorbed energy by azo-dye during exposure translates the azo-dye molecules from their trans-

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longitudinal state to an unstable cis-excited state. As the absorbed energy in the trans-state is proportional the orientation of azo-dye axis of molecular with respect to polarization of incident light, since the anisotropic property of azo-dye, the aligning power of azo-dye is slow while the minimal energy configuration of azo-dye satisfies at the condition of perpendicular to light polarization with azo-dye molecules axis [149].

The orientation of nematic liquid crystal is controllable by doping appropriate azo-dye. Those azo-dyes as photoexcited dye molecules are found to exert a torque to reorient the director axis liquid crystals through intermolecular interactions with host molecules [150]. The dynamic property of azo-dye-doped liquid crystals in holography has been studied by A. G. Chen etc in early time [151, 152]. The photo alignment property of azo-dye in liquid crystals has been investigated by a lot of people in recently [153-156]. As a good candidate for polarization related optical devices, azo-dye-doped nematic liquid crystal has been reported to involve in fabrication of polarization Fresnel lens [157, 158], polarization grating [159-161], vertical photonic crystals [162], polarization rotators [163], and photonic crystal fiber devices [164].

4.2 Two-Order Binary Circular Dammann Grating Based On

Azo-Dye-Doped Liquid Crystals

Previously reported CDGs were usually fabricated by materials such as glass, which could be used only for one fixed laser wavelength, limiting the application range and flexibility. In our experiment, a broad band polarization-dependent circular Dammann grating was generated from an azo-dye-doped nematic liquid crystal cell. A binary phase liquid crystal two-order CDG zone plate consisted of an odd zone with twisted nematic (TN) LC structure and an

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even zone with homogenous LC structure was fabricated through a multi-exposure photo-alignment process. The maximum 0th and 1st/2nd diffraction orders of CDG can be separately achieved by changing the analyzer's polarization direction, thus leading to a polarization-dependent tunable CDG. The CDG using azo-dye-doped LC cell can be used to generate diffractions by lasers in a broad wavelength range, hence expanding possible device applications.

Using the optimization process introduced above, the numerical simulation results for small order CDG, e.g. one-order to three-order, were given in table 1 [137].

Table 1 Numerical Results of Binary Phase (0, π) CDGs.

Order Number	Transition Number (N)	Transition Points	Efficiency	Uniformity
1	2	0.5	0.810	
2	4	0.1634	0.652	0.0001
3	6	0.1677, 0.4203	0.671	0.0012
4	8	0.1301, 0.2492, 0.4643	0.710	0.0007

Figure 4-2 (a) Cross section of a two-order binary phase (0, π) CDG. (b) Theoretical normalized intensity of a two-order CDG.

Figure 4-3 Two-order binary phase CDG pattern transferred onto a photo-mask.

For the case of two-order CDG, with profile shown in Figure 4-2(a), where $\{r_1=0.1634, r_2=0.5, r_3=0.8366, r_4=1\}$ are the normalized phase transitions in a period length and chosen from table 1 directly, the corresponding theoretical spectral intensity in the far field is presented in Figure 4-2(b). The calculated coefficients c_n are given by

$$c_n = \frac{2}{n\pi} \left[-2 \cos(2\pi n x_1) + (-1)^{n+2} + 1 \right]. \quad (4.16)$$

where, $x_1=r_1=0.1634$. The designed two-order binary phase CDG pattern was then transferred to a photo-mask with a period of $T=200 \mu\text{m}$, a total aperture of 8 mm, and total periodic number of $m=20$, as depicted in Figure 4-3. The photo mask was fabricated based on a glass substrate with a layer of chrome on one side. During the process of pattern fabrication, some portion of chrome portion, corresponding to the white part in Figure 4-3, was removed either by direct-written on e-beam or laser system or manufactured from reticles using photorepeater system, the photo mask was fabricated by Photonik Singapore Pte Ltd based on the specific design with detail in Figure 4-3.

4.3 Experimental Process of Fabrication

The materials used herein consisted of 99 wt% liquid crystal, E7 ($n_o = 1.5216$ and $n_e = 1.7462$) from Merck, and 1 wt% azo-dye, Methyl Red (MR) from Aldrich. Two indium-tin-oxide (ITO) coated glass slides were used to assemble an empty cell with a gap of $d=30 \mu\text{m}$. The dye-doped LC mixture was then infiltrated into an empty cell. As both glass substrates were not rubbed, the alignment of LC in cell was initially random.

The LC molecules in the cell can be reoriented by linearly polarized light in the absorption band of azo-dye. When excited by light in the absorption

spectrum, the azo-dye in LC sample would undergo *trans-cis* isomerization, followed by molecular reorientation, diffusion and adsorption on the ITO glass surface. The adsorbed dyes then align liquid crystals by forcing the LC molecules to reorient along the direction of azo-dye's long axis.

Figure 4-4 Schematic fabrication process of azo-dye-doped LC circular Dammann grating.

In our experiment, a diode-pumped linearly polarized solid state laser light at a wavelength of 532 nm, which is in the absorption band of MR dye (410-550 nm, with its peak around 500 nm), was used to irradiate the dye-doped LC sample for photo-aligning LC molecules in the cell. The absorption of MR dye at 532 nm is high, which is about 70% of that at peak absorption wavelength of 500nm. The photo alignment process for fabricating circular Dammann grating on the azo-dye-doped liquid crystal sample consists of two steps. In the first step, as shown in Figure 4-4(a), glass substrates 1 was illuminated by the laser light with an intensity level of 120 mW/cm^2 for 3

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minutes with linear polarization along x axis. In the second step, as shown in Figure 4-4(b), similar treatment is used on glass substrates as the first step. Then the MR dye on substrate aligned LC to be perpendicular to the incident light polarization, leading to a parallel alignment along y axis of LC on both substrates and therefore formation of a homogeneous LC layer structure, as shown in Figure 4-4 (c). Figure 4-5 shows the absorption spectrum of the azo-dye doped LCs after the first alignment step, which confirmed the alignment of azo-dye and LC molecules. The absorption intensity of perpendicular polarized light is smaller than that of parallel polarized light with respect to the director axis of LC (y axis as shown in Figure 4-4 (c)), where the strong anisotropic absorption indicates that the azo-dye molecules are well aligned with the LC.

In the third step, as shown in Figure 4-4(d), the glass substrate 2, covered by the photo mask with circular Dammann grating pattern, was illuminated by laser with linear polarization along y axis. The odd circle/rings of the CDG are transparent and the even rings are opaque. During the third step, the MR dye on substrate 2 reoriented the LC again and a twisted nematic (TN) structure for the odd zone was achieved finally, while the homogenous structure of the even zone remained unchanged, as shown in Figure 4-4(e). The adsorption-induced twist angle, defined as the angle between the original LC directors direction and the reoriented LC molecule directors by azo-dye, is dependent on the exposure time and pumping intensity of illumination light [163]. In our experiment, for demonstrating a more general case, a 60° TN structure for odd circle/rings of CDG zone was firstly obtained by using a laser intensity of 120 mW/cm^2 and exposure time of 2 minutes for the second illumination in Figure 4-4(d). A larger twist angle TN structure (e.g. 90° TN)

could be obtained by lengthening the exposure time or increasing the illumination intensity. Such multi-exposure photo-alignment process provides a simple way to align LC in cell as no rubbing is needed.

Figure 4-5 Absorption spectra of nematic liquid crystal (E7) doped with azo dye MR (1%). $A_{\perp}$ and $A_{\parallel}$ represent the absorbance of the azo-dye doped LC cell (Figure 4-4(c)), after the first alignment step, measured by light polarization perpendicular and parallel to the director of LC (y axis) respectively.

For odd zone of cell with θ^0 TN structure (where θ is the twist angle between LC directors on substrate 1 and 2) as presented in Figure 4-4(e), if light with linear polarization along y axis (parallel to the LC director on substrate 1) is incident from substrate 1 side, the θ^0 TN structure will rotate the polarization of light by $\sim \theta^0$ after it passes through the cell when the Mauguin's condition is satisfied ($\Delta n d \gg \lambda$, where $\Delta n = n_e - n_o$ and λ is wavelength of incident light) [165], which means the polarization of output light passing through odd zone will be rotated to be parallel to the LC director on substrate 2. In our

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experiment, the Mauguin's condition was satisfied and θ was equal to 60° . For even zone with homogeneous structure, the polarization of incident light would not be changed after it passes through the cell, when light with polarization along y axis is incident from substrate 1.

When the LC cell sample was examined under an optical microscope with two polarizers, the white light source would pass through the polarizer 1(P1), substrate 1, LC cell, substrate 2, and polarizer 2 (P2) in sequence in our experiment. The direction of polarizer 1 was set to be parallel to the LC director on substrate 1, both along the y axis. The obtained microscope images of photo-aligned azo-dye-doped LC with circular Dammann grating pattern are shown in Figure 4-6 where Figure 4-6(a) and 4-6(b) represent images captured with polarizer-polarizer angles of 30° and 90° , corresponding to a completely dark odd zone and a completely dark even zone respectively. The directions of polarizer 1 and polarizer 2 for each case are drawn below each image. For the odd zone, a completely darkened image appeared only when polarizer 2 had a 30° angle with polarizer 1 as shown in Figure 4-6(a), indicated that the LC director (D) on substrate 2 should be perpendicular to polarizer 2 and has a 60° angle with the y axis as shown in the left part of Figure 4-6(c). Therefore, a 60° TN structure can be confirmed in the odd circle/rings zone. Similarly, for the even rings zone, a completely darkened image appeared only when polarizer 2 was perpendicular to polarizer 1 as shown in Figure 4-6(b), confirming a parallel alignment LC structure. The schematic of fabricated LC cell with CDG pattern is shown in Figure 4-7. The LC directors will keep unchanged in even zone and show 60° TN structure in odd zone.

Figure 4-6 Optical microscope images of the photo-aligned azo-dye-doped LC cell with circular Dammann grating pattern in the cases of (a) 30° angle of polarizer-polarizer, and (b) crossed polarizer-polarizer. Solid line and dashed line represent polarizer 1 and polarizer 2, respectively. The scale bars are $200\text{ }\mu\text{m}$. (c) The directions of LC directors on substrate 1 and 2 are shown for odd and even rings zones, respectively.

Figure 4-7 Schematic of fabricated LC cell with CDG pattern. The LC directors will keep unchanged in even zone and show 60° TN structure in odd zone. Letters *e* and *o* are used to represent the even and odd zone respectively.

For generating CDG diffraction, the azo-dye-doped LC cell sample recorded with CDG pattern was illuminated by a collimated He-Ne laser (633 nm) with a beam diameter of about 1 cm and intensity of 0.2 mW/cm^2 to generate the CDG. The absorption of MR at 633nm is very low, which is around 10% of that at peak absorption wavelength of 500nm. A polarizer was used to ensure the polarization of incident light was parallel to the LC director on substrate 1 (both along y axis). A spherical lens ($f=20 \text{ cm}$) placed right behind the LC cell sample was used to perform the Fourier transform of the CDG pattern. An analyzer was used to verify the image polarization. The circular diffraction pattern was obtained at the focal plane of the Fourier-transform lens, and then captured by a charge-coupled-device (CCD) camera.

4.4 Polarization Dependent Property

Figure 4-8 shows the polarization dependent property of the azo-dye-doped LC circular Dammann grating recorded by CCD. The polarization of the incident light, parallel to the LC directors on substrate 1 in even zone, would not change because the even zone has a homogeneous LC structure, while the polarization of the incident light through the odd zone is rotated by $\sim 60^\circ$ because the odd zone has 60° TN LC structure. That is called waveguiding effect [165] in TN LC structure. As a result, the angle between the polarizations of output light from even and odd zones is around 60° . Figure 4-8 presents the observed images recorded by a CCD, with (a) no analyzer (b) 30° , (c) 15° , (d) 0° , (e) -15° , (f) -30° , (g) -45° , and (h) -60° of analyzer-polarizer angle, where the 0° is along y axis. Corresponding output light polarization from even (E_{even}) and

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odd (E_{odd}) zone, polarization (P), and analyzer (A) directions are indicated below each image, respectively.

Figure 4-8(a) shows the observed image without an analyzer, where both of the 0^{th} and $1^{\text{st}}/2^{\text{nd}}$ diffraction order corresponding to light spot and circular Dammann grating two-equal intensity rings existed. Figure 4-8(b) and 4-8(h) show the observed images with the maximum 0^{th} and $1^{\text{st}}/2^{\text{nd}}$ diffraction order, where the analyzer was set to 30° and -60° , comparing to the polarizer direction (y axis), respectively. The reason behind this observed image can be found as follow. Assuming the incident light has a polarization state of parallel to 0° defined in Figure 4-8, after passing through the CDG, the polarization of output light in even zone kept unchanged while that in odd zone became parallel to 60° defined in Figure 4-8. When the analyzer was set along the direction of 30° defined in Figure 4-8, the electric field components of $\vec{E}_{\text{even}}$ and $\vec{E}_{\text{odd}}$ along the direction of analyzer are equal to each other with both the same amplitude and phase, leading to a zero phase difference of output light from even and odd zones and finally the 0^{th} diffraction order maximum.

Figure 4-8 The observed images recorded by a CCD. (a) Without an analyzer, and with (b) 30° , (c) 15° , (d) 0° , (e) -15° , (f) -30° , (g) -45° , and (h) -60° of analyzer-polarizer angle, where the 0° is along y axis. Corresponding output light polarization from even (E_{even}) and odd (E_{odd}) zones, polarization (P), and analyzer (A) directions are shown below each image, respectively.

In contrast, when the analyzer was set along -60° defined in Figure 4-8, the electric field components of $\vec{E}_{\text{even}}$ and $\vec{E}_{\text{odd}}$ along the direction of analyzer only have the same amplitude but a π phase difference, leading to a π phase difference of output light from even and odd zone and finally the desired CDG image with the $1^{\text{st}}/2^{\text{nd}}$ diffraction order maximum. If the analyzer was set between 30° and -60° , the obtained images would be in one of the intermediate status with both 0^{th} and $1^{\text{st}}/2^{\text{nd}}$ orders, and these corresponding images are given in Figure 4-8(c)-(g). Therefore, by adjusting the analyzer's direction, we can

gradually change the output circular Dammann grating image, which makes it possible to use as a polarization dependent tunable optical element.

From Figure 4-8(h), we can see that two equal-intensity rings are produced from our sample as predicted, the unwanted zero order in CDG image was due to its high sensitivity to fabrication errors, where a very bright zero order can be produced even when a small phase error existed. However, this did not prevent demonstrating the utility of the polarization-dependent azo-dye-doped LC CDG. The splitting of the main intensity maximum are due to the property of binary phase CDG and could be eliminated by a four-phase level CDG [137].

Figure 4-9 Cross section of normalized intensity of the two-order binary CDG obtained in experiment.

Figure 4-9 shows the normalized intensity of the two-order binary CDG given in Figure 4-8(h), where ρ represents the distance from the center of CDG in the polar coordinates. The splitting of the main intensity maximum, as discussed previously by S. Zhao et al., are due to the property of binary phase CDG and could be eliminated by a four-phase level CDG [137]. The radii of produced intensity rings can be calculated from $q=\rho/\lambda f=n/T$, and the theoretical

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radius of the 1st and 2nd order rings on image plane are $\rho_1 = \lambda f/T = 633 \text{ } \mu\text{m}$, and $\rho_2 = 2\lambda f/T = 1266 \text{ } \mu\text{m}$, respectively, where $\lambda = 633 \text{ nm}$, $f = 20 \text{ cm}$, and $T = 200 \text{ } \mu\text{m}$ in our experiment. The obtained experimental ρ_1 and ρ_2 were around $606 \text{ } \mu\text{m}$ and $1263 \text{ } \mu\text{m}$, respectively, slightly smaller than what is predicted, which was due to the errors of radii's position in binary phase CDG [137]. Therefore, our experimental results are quite consistent with the simulation.

It is worth noting that, any laser 1) satisfying Mauguin's condition, and 2) that is sufficiently far away from MR azo-dye absorption peak or even near the absorption peak but then with low intensity, could be used to generate the polarization-dependent CDG diffraction owing to the light-rotating (waveguiding) property of TN structure in odd zone. This broad band property expands the CDG application range which is of great advantage comparing to previously reported CDGs operating at only one fixed working wavelength.

Additionally, it is worth noticing that, the rotation angle of azo-dye doped liquid crystal under local polarization of incident laser beam depends on the intensity and exposure time of laser light as mentioned above. In our experiment, a 55° TN structure for odd circle/rings of CDG zone was obtained by the same laser intensity of 120 mW/cm^2 but shorter exposure time of 1.5 minutes for the second illumination in Figure 4-4(d), while keeping all else unchanged. The examination optical images under optical microscopy with cross polarizers and corresponding generated polarization dependent CDG were shown in Figure 4-10 and Figure 4-11 respectively.

Figure 4-10 Optical microscope images of the photo-aligned azo-dye-doped LC cell with circular Dammann grating pattern in cases of (a) The directions of LC director on substrate 1 and 2 for odd and even rings zones, respectively, (b) -35° angle of polarizer-polarizer, (c) 27.5° angle of polarizer-polarizer and (d) crossed polarizer-polarizer. Solid line and dash line represent polarizer 1 and polarizer 2 respectively.

Figure 4-1 The observed images recorded by a CCD. (a) Without analyzer, (b) with 27.5° , (c) -62.5° of analyzer-polarizer angle. Corresponding output light polarization from even (E_{even}) and odd (E_{odd}) zone, polarization (P), and analyzer (A) directions are shown below each image, respectively.

4.5 Conclusion

In conclusion, we have generated a polarization-dependent circular Dammann grating diffraction from an azo-dye-doped LC cell. A simple multi-exposure photo-aligned process was used to fabricate a binary phase liquid crystal CDG zone plate that consisted of an even zone with homogenous LC structure and an odd zone with 60° and 55° TN LC structure. The maximum 0^{th} and $1^{\text{st}}/2^{\text{nd}}$ diffraction orders of CDG can be separately achieved by changing the analyzer's polarization direction. If the chosen analyzer's direction leads to a zero phase difference in output light from even and odd zones, the maximum 0^{th} diffraction order will be achieved. In contrast, if the chosen analyzer's direction leads to a π phase difference in output light from even and odd zones, the maximum 1^{st} diffraction order will be produced. The CDG diffraction from our azo-dye-doped LC cell can be generated by lasers within a broad wavelength band, which expands possible applications.

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5.1 Introduction

5.1.1 Photonic Crystals

Photonic crystals are periodic optical structures, which is designed for controlling the movement of photon. They are similar to semiconductors with periodic structures in atomic scale. Generally, photonic crystals contain repeated internal materials with high and low dielectric constant. Photons propagate through this structure - or not - depending on the relationship of their wavelength and photonic crystal structure. Lord Rayleigh studied the one-dimensional periodic structure of multi-layer dielectric stacks in 1887, by showing that such systems have a one-dimensional photonic band-gap, a spectral range of large reflectivity, known as a stop-band.

Figure 5-1 Examples of 1D, 2D and 3D photonic crystal structures [166].

In 1987, Yablonovith [167] and John [168] published two milestone papers both discussing high dimensional periodic optical structures. Yablonovith discussed a three-dimensionally periodic dielectric structure with an electromagnetic band gap, where spontaneous emission can be rigorously forbidden [167]; John proposed a mechanism for strong Anderson localization of photons in carefully prepared disordered dielectric superlattices with an everywhere real positive dielectric constant [168]. The example of 1D, 2D and 3D photonic crystal structures are shown in Figure 5-1. Two different colors represent for two materials with different dielectric constants, respectively.

Photonic crystals are attractive optical materials useful for controlling and manipulating the flow of light. The study of photonic crystal is governed by Bloch theorem and Maxwell's equation. To design photonic crystal structure, it is important to engineer the location or size of the bandgap in photonic band structure. The two most popular numerical methods for calculating photonic band structures are plane wave method (PWM) [169] and Finite Difference Time Domain (FDTD) [170], other numerical methods include transfer matrix method [166, 171], scattering matrix method [172], finite element method [173, 174], and KKR method [175].

5.1.2 Plane Wave Expansion Method

Plane wave method (PWM), a frequency domain simulation method, was used in calculation of band structure of photonic crystals in 1990s and treated as a breakthrough in photonic crystal band structure calculation. It is a full vector method to deal with macroscopic electromagnetic problems by adopting periodic boundary. This concept may extend to even a lot of non-periodic problems applying supercell conception once the effect at the boundary

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can be treated small enough. Plane wave method is quite useful in calculating band structure and mode distribution of photonic crystals with periodic structures. We will start from Maxwell equation to deduce the plane wave method applied in solving photonic crystals band structures.

The full macroscopic Maxwell's equations can be expressed as [176]:

$$\begin{aligned}\nabla \cdot \mathbf{D}(\mathbf{r}, t) &= \rho_0(\mathbf{r}, t) \\ \nabla \cdot \mathbf{B}(\mathbf{r}, t) &= 0 \\ \nabla \times \mathbf{H}(\mathbf{r}, t) &= \frac{\partial \mathbf{D}(\mathbf{r}, t)}{\partial t} + \mathbf{J}_0(\mathbf{r}, t) \\ \nabla \times \mathbf{E}(\mathbf{r}, t) &= -\frac{\partial \mathbf{B}(\mathbf{r}, t)}{\partial t}\end{aligned}\tag{5.1}$$

where $\mathbf{E}$ and $\mathbf{H}$ represent the electric field and magnetic field respectively, $\mathbf{D}$ and $\mathbf{B}$ represent the electric flux density and magnetic flux density respectively, ρ_0 is free charge density, and $\mathbf{J}_0$ is free electric current density.

Considering electromagnetic wave in materials with [176]:

1. Source free space: $\rho_0(\mathbf{r})=0$, $\mathbf{J}_0(\mathbf{r})=0$;
2. Lossless medium: $\varepsilon(\mathbf{r})$ is real in our interested region;
3. Linear and time invariant: plane wave theory is available;
4. Magnetic uniform: $\mu(\mathbf{r})=\text{constant}$.

Taking the forms of $\mathbf{E}$ and $\mathbf{H}$ oscillating sinusoidally with time, we can write [176]:

$$\begin{aligned}\mathbf{E}(\mathbf{r}, t) &= \mathbf{E}(\mathbf{r})e^{j\omega t} \\ \mathbf{H}(\mathbf{r}, t) &= \mathbf{H}(\mathbf{r})e^{j\omega t}\end{aligned}\tag{5.2}$$

Using relationship of $\mathbf{D}(\mathbf{r}) = \varepsilon_0 \varepsilon(\mathbf{r}) \mathbf{E}(\mathbf{r})$ and $\mathbf{B}(\mathbf{r}) = \mu_0 \mathbf{H}(\mathbf{r})$, noted that

$\partial / \partial t \rightarrow j\omega$, we can obtain [176]:

$$\begin{aligned}
\nabla \cdot \varepsilon(\mathbf{r}) \mathbf{E}(\mathbf{r}) &= 0 \\
\nabla \cdot \mathbf{H}(\mathbf{r}) &= 0 \\
\nabla \times \mathbf{H}(\mathbf{r}) &= j\omega \varepsilon_0 \varepsilon(\mathbf{r}) \mathbf{E}(\mathbf{r}) \\
\nabla \times \mathbf{E}(\mathbf{r}) &= -j\omega \mu_0 \mathbf{H}(\mathbf{r})
\end{aligned} \tag{5.3}$$

where ε_0 and μ_0 are free space electrical permittivity and free space magnetic permeability respectively, $\varepsilon(\mathbf{r})$ is relative permittivity of medium. $\nabla \cdot \mathbf{H} = \nabla \cdot \mathbf{D} = 0$ indicates: a) There is no charge or current source; b) $\mathbf{H}$ and $\mathbf{D}$ are built up of transverse plane waves, where $\mathbf{k} \cdot \mathbf{H} = 0$ and $\mathbf{k} \cdot \mathbf{D} = 0$, and continues everywhere. But $\mathbf{E}$ is not necessarily transverse and continuous.

From Eq. (5.3), we can get [176]:

$$\nabla \times \left(\frac{1}{\varepsilon(\mathbf{r})} \nabla \times \mathbf{H}(\mathbf{r}) \right) = \frac{\omega^2}{c^2} \mathbf{H}(\mathbf{r}). \tag{5.4}$$

This is a standard eigen-value problem, which can be written as $\Theta \mathbf{H} = \Lambda \mathbf{H}$. The

operator $\Theta = \nabla \times \frac{1}{\varepsilon} \nabla \times$ has several important properties [170]:

- 1) It is a linear operator: if $\mathbf{H}_1$ and $\mathbf{H}_2$ are two different modes of the system, then the combination $\mathbf{H}_1 + \mathbf{H}_2$ is also a mode of the system.
- 2) It is a Hermitian operator, which satisfy the inner product $(f, \Theta g) = (\Theta f, g)$ by definition and has two important properties:
 - Firstly, it has only real eigen values, because it is a Hermitian operator and $\varepsilon(\mathbf{r})$ is real everywhere.
 - Secondly, any two different modes are orthogonal to each other, which means $(\mathbf{H}_i, \mathbf{H}_j) = \delta_{ij}$ assuming the magnetic fields are normalized.
- 3) Scaling. If the same dielectric function scales by a factor of a , $\varepsilon'(\mathbf{r}) = \varepsilon(\mathbf{r}/a)$, the eigen frequency will be scaled by factor of a , $\omega' = \omega/a$. This is

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the reason why the analysis of band structure of photonic crystal can be applied to different frequency region such as optical or microwave region.

Once the $\mathbf{H}$ has been solved, we can obtain $\mathbf{E}$ through following expression [176]:

$$\mathbf{E}(\mathbf{r}) = \frac{1}{j\omega\epsilon(\mathbf{r})} \nabla \times \mathbf{H}(\mathbf{r}). \quad (5.5)$$

Here we don't use the equation with replacement of $\mathbf{D}$ or $\mathbf{E}$, because the operators for $\mathbf{D}$ and $\mathbf{E}$ are not Hermitian and do not possess these properties and increase the complexity of equations.

Bloch theorem is often applied in the case of infinite periodic structures. The Bloch theorem indicates that a plane wave in an infinite periodic structure will be modulated by the periodicity [176]. In photonic crystal, the $\epsilon(\mathbf{r})$ is a periodic function, therefore the magnetic field $\mathbf{H}$ can be expressed as [176]:

$$\begin{aligned} \mathbf{H}(\mathbf{r}) &= e^{i\mathbf{k} \cdot \mathbf{r}} h(\mathbf{r}) \hat{\mathbf{e}}_{\mathbf{k}} \\ h(\mathbf{r}) &= h(\mathbf{r} + \mathbf{R}_l) \end{aligned} \quad (5.6)$$

where $\mathbf{R}_l$ is an arbitrary lattice vector, $\hat{\mathbf{e}}_{\mathbf{k}}$ is a unit vector perpendicular to the vector $\mathbf{k}$ and parallel to the $\mathbf{H}$ vector to reflect the transversal property of $\mathbf{H}$.

The dielectric function $\epsilon(\mathbf{r})$ is periodic in space of $\mathbf{r}$, it can be given by [176]:

$$\epsilon(\mathbf{r}) = \epsilon(\mathbf{r} + \mathbf{R}_l), \quad (5.7)$$

where,

$$\mathbf{R}_l = l_1 \mathbf{a}_1 + l_2 \mathbf{a}_2 + l_3 \mathbf{a}_3, \quad (5.8)$$

$\mathbf{R}_l$ is an arbitrary lattice vector, l_1, l_2, l_3 are integers, and $\mathbf{a}_1, \mathbf{a}_2, \mathbf{a}_3$ are the basis lattice vectors to describe the periodic lattice.

Any periodic function can be expanded using Fourier series [176]:

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$$\varepsilon(\mathbf{r}) = \sum_{\mathbf{G}_i} \varepsilon(\mathbf{G}_i) e^{i\mathbf{G}_i \cdot \mathbf{r}}, \quad (5.9)$$

$$\frac{1}{\varepsilon(\mathbf{r})} = \sum_{\mathbf{G}_i} \varepsilon^{-1}(\mathbf{G}_i) e^{i\mathbf{G}_i \cdot \mathbf{r}}. \quad (5.10)$$

Since $h(\mathbf{r})$ has similar periodic property as $\varepsilon(\mathbf{r})$ [176]:

$$h(\mathbf{r}) = \sum_{\mathbf{G}_i} h(\mathbf{G}_i) e^{i\mathbf{G}_i \cdot \mathbf{r}}. \quad (5.11)$$

$H(\mathbf{r})$ can be expanded as [176]:

$$H(\mathbf{r}) = \hat{e}_k e^{i\mathbf{k} \cdot \mathbf{r}} \sum_{\mathbf{G}_i} h(\mathbf{G}_i) e^{i\mathbf{G}_i \cdot \mathbf{r}} = \sum_{\mathbf{G}_i, \lambda} h(\mathbf{G}_i, \lambda) e^{i(\mathbf{k} + \mathbf{G}_i) \cdot \mathbf{r}} \hat{e}_{\lambda, \mathbf{k} + \mathbf{G}_i}. \quad (5.12)$$

Here $\mathbf{G}_i$ represents an arbitrary spatial frequency which is called reciprocal lattice vector can be expressed as [176]:

$$\mathbf{G}_i = h_1 \mathbf{b}_1 + h_2 \mathbf{b}_2 + h_3 \mathbf{b}_3, \quad (5.13)$$

where $\mathbf{b}_1, \mathbf{b}_2, \mathbf{b}_3$ are basic vectors in the reciprocal space ; h_1, h_2, h_3 are integers; λ represents 1 and 2. In Eq. (5.12), the transverse property of $H(\mathbf{r})$ is used to decompose the wave into sum of a set of plane wave, which is the core of this method. $h(\mathbf{G}_i, \lambda)$ is the component amplitude along the unit vector, $\hat{e}_{1, \mathbf{k} + \mathbf{G}_i}$ and $\hat{e}_{2, \mathbf{k} + \mathbf{G}_i}$ are perpendicular to each other and are both perpendicular to $\mathbf{k} + \mathbf{G}_i$ and $h(\mathbf{G}_i, \lambda)$. Therefore vectors of $\mathbf{k} + \mathbf{G}_i$, $\hat{e}_{1, \mathbf{k} + \mathbf{G}_i}$, $\hat{e}_{2, \mathbf{k} + \mathbf{G}_i}$ forms a Cartesian triad [176]:

$$\begin{aligned} (\mathbf{k} + \mathbf{G}_i) \cdot \hat{e}_\lambda &= 0 \\ \hat{e}_1 \cdot \hat{e}_2 &= 0 \end{aligned} \quad (5.14)$$

From solid-state physics, all the lattice vectors $\mathbf{R}_l$ from the lattice in the $\mathbf{r}$ -space, while all the reciprocal lattices vectors $\mathbf{G}_i$ form the reciprocal lattice in the reciprocal spacem [176]. Those two sets of vectors are orthogonal to each other with following relationship [176]:

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$$\mathbf{b}_i \cdot \mathbf{a}_j = 2\pi\delta_{ij} \quad (5.15)$$

$$\mathbf{b}_1 = 2\pi \frac{\mathbf{a}_2 \times \mathbf{a}_3}{\mathbf{a}_1 \cdot \mathbf{a}_2 \times \mathbf{a}_3}, \mathbf{b}_2 = 2\pi \frac{\mathbf{a}_3 \times \mathbf{a}_1}{\mathbf{a}_1 \cdot \mathbf{a}_2 \times \mathbf{a}_3}, \mathbf{b}_3 = 2\pi \frac{\mathbf{a}_1 \times \mathbf{a}_2}{\mathbf{a}_1 \cdot \mathbf{a}_2 \times \mathbf{a}_3} \quad (5.16)$$

Using Eq. (5.10), Eq. (5.12), and Eq. (5.4), after complicated calculation [176], we can obtain following [177]:

$$\sum_{\mathbf{G}'} |\mathbf{k} + \mathbf{G}| |\mathbf{k} + \mathbf{G}'| \varepsilon^{-1}(\mathbf{G} - \mathbf{G}') \begin{bmatrix} \hat{\mathbf{e}}_2 \cdot \hat{\mathbf{e}}'_2 & -\hat{\mathbf{e}}_2 \cdot \hat{\mathbf{e}}'_1 \\ -\hat{\mathbf{e}}_1 \cdot \hat{\mathbf{e}}'_2 & \hat{\mathbf{e}}_1 \cdot \hat{\mathbf{e}}'_1 \end{bmatrix} \begin{pmatrix} h'_1 \\ h'_2 \end{pmatrix} = \frac{\omega^2}{c^2} \begin{pmatrix} h_1 \\ h_2 \end{pmatrix} \quad (5.17)$$

This is a standard eigen-value problem and it can be represented in the matrix form [177]:

$$\begin{bmatrix} M_1 & M_2 \\ M_3 & M_4 \end{bmatrix} \begin{pmatrix} h_1(\mathbf{G}') \\ h_2(\mathbf{G}') \end{pmatrix} = \frac{\omega^2}{c^2} \begin{pmatrix} h_1(\mathbf{G}) \\ h_2(\mathbf{G}) \end{pmatrix} \quad (5.18)$$

If N plane waves are used in equation, there will be a $2N$ linear equation group exist. For 2D photonic crystal case, we assume the dielectric constants along z -direction are invariant. For in-plane propagation of x - y plane, $k_z=0$, thus wave vectors $\mathbf{k}$ and $\mathbf{G}$ are in x - y plane, and $\mathbf{k} + \mathbf{G}$ is also in x - y plane. Setting one of the unit vectors is constantly to $\hat{\mathbf{e}}_{2,\mathbf{k}+\mathbf{G}} = \hat{\mathbf{z}}$, the other unit vector $\hat{\mathbf{e}}_{1,\mathbf{k}+\mathbf{G}}$ is also in the x - y plane. Using $\hat{\mathbf{e}}_1 = \langle \cos\theta, \sin\theta, 0 \rangle, \hat{\mathbf{e}}_2 = \langle 0, 0, 1 \rangle$, we can obtain[177]:

$$\begin{bmatrix} \hat{\mathbf{e}}_2 \cdot \hat{\mathbf{e}}'_2 & -\hat{\mathbf{e}}_2 \cdot \hat{\mathbf{e}}'_1 \\ -\hat{\mathbf{e}}_1 \cdot \hat{\mathbf{e}}'_2 & \hat{\mathbf{e}}_1 \cdot \hat{\mathbf{e}}'_1 \end{bmatrix} = \begin{bmatrix} 1 & 0 \\ 0 & \cos(\theta - \theta') \end{bmatrix} \quad (5.19)$$

The $2N$ by $2N$ equation group is decoupled into two equation groups: TE and TM modes. One group only has $\mathbf{H}$ components along $\hat{\mathbf{e}}_1$ direction, and no components along $\hat{\mathbf{e}}_2 = \hat{\mathbf{z}}$, which is called TM wave. The other group only

has E components along $\hat{e}_1$ direction, and no components along $\hat{e}_2 = \hat{z}$, which is called TE wave. Eq. (5.17) can be rewritten as [177]:

$$\text{TM: } \sum_{\mathbf{G}'} |\mathbf{k} + \mathbf{G}| |\mathbf{k} + \mathbf{G}'| \varepsilon^{-1}(\mathbf{G} - \mathbf{G}') h_1(\mathbf{G}') = \frac{\omega^2}{c^2} h_1(\mathbf{G}) \quad (5.20)$$

$$\text{TE: } \sum_{\mathbf{G}'} (\mathbf{k} + \mathbf{G}) \cdot (\mathbf{k} + \mathbf{G}') \varepsilon^{-1}(\mathbf{G} - \mathbf{G}') h_2(\mathbf{G}') = \frac{\omega^2}{c^2} h_2(\mathbf{G}) \quad (5.21)$$

Each of above is a group of N linear equation when N plane waves are used. For each specific wave vector of $\mathbf{k}$, the eigen-value results the frequencies ω of the eigen-modes. Using N plane waves, $2N$ (TE for N and TM for N) discrete frequencies was obtained for each $\mathbf{k}$ -point. These frequencies are sorted in ascending order while labeled as 1 to $2N$. One band of photonic crystals structure is formed by all the eigen-frequencies with the same order for all $\mathbf{k}$ -vectors in first irreducible Brillouin zone (IBZ) [176].

The wave vector $\mathbf{k}$ can be restricted in the First Brillouin zone for the periodicity in the reciprocal space according to Bloch theorem. Using the symmetry of the first Brillouin zone, the eigen-frequencies for those $\mathbf{k}$ -vectors along the edge of the IBZ can be calculated, as those frequencies for the $\mathbf{k}$ -vectors locating inside the IBZ will locate inside the band. The concepts of physics such as band structure, lattice vectors, reciprocal vectors, basic vectors, the first Brillouin zone, and irreducible Brillouin zone are all taken from solid-state physics.

5.1.3 Two-Dimensional Photonic Crystal Lattice Structure

As the photonic crystal are commonly formed by confined medium locating at periodic points with another background medium containing the

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confined medium [176], normally, the confined medium is called the “atom” for convenience. There are essentially only two kinds of 2D lattice structures. One is the orthogonal lattice, in which the two basis vectors are orthogonal, e.g. square lattice; another is the oblique lattice, in which the two basis vectors are not orthogonal, e. g. triangle structure, honeycomb lattice and Kagomé lattice. Figure 5-2 to Figure 5-5 shows the lattice structure of square, triangle, honeycomb, and Kagomé respectively. Table 2 shows parameters of various 2D lattices.

Figure 5-2 Two dimensional square lattice, unit cell and Brillouin zone. Left: unit cell and basis lattice vector. Right: The first Brillouin zone and the irreducible Brillouin zone. Γ , M , X represent high symmetry points [176].

Figure 5-3 Two dimensional triangle lattice, unit cell and Brillouin zone. Left: unit cell and basis lattice vector. Right: The first Brillouin zone and the irreducible Brillouin

zone. Γ , M, X represent high symmetry points [176].

Figure 5-4 Two dimensional honeycomb lattice, unit cell. The first Brillouin zone and the irrduciable Brillouin zone are same to Figure 5-3[176].

Figure 5-5 Two dimensional Kagomé lattice, unit cell. The first Brillouin zone and the irrduciable Brillouin zone are same to Figure 5-3[176].

Considering a unit cell with an area of A , the circular atom which is located in the cente, is inversion symmetric, ensuring the Fourier transform is real. If the unit cell is represented by function of $f(\mathbf{r})$ [176],

$$f(\mathbf{r}) = \begin{cases} \varepsilon_a & \dots r < R \\ \varepsilon_b & \dots r > R \end{cases} = \varepsilon_b + \begin{cases} \varepsilon_a - \varepsilon_b, & r < R \\ 0, & r > R \end{cases} \quad (5.22)$$

The Fourier transform is given by [176]:

$$F(\mathbf{k}) = \frac{1}{A} \iint_A f(\mathbf{r}) e^{-i\mathbf{k} \cdot \mathbf{r}} d\mathbf{r} \quad (5.23)$$

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After complicated deduce, we can obtain the Fourier transform for square and triangle lattice [176]:

$$\begin{aligned} F(0) &= \varepsilon_b + (\varepsilon_a - \varepsilon_b)f & \mathbf{G} &= 0; \\ F(\mathbf{G}) &= 2(\varepsilon_a - \varepsilon_b)f \frac{J_1(GR)}{GR} & \mathbf{G} &\neq 0 \end{aligned} \quad (5.24)$$

where J_1 represents the first order Bessel function.

Table 2 Various 2D lattice, lattice constant, a , is defined as the distance between the two closest “atoms” [176].

Lattice Type	Basic Vector	Reciprocal Vectors	Position of “Atom”
Square Lattice	$a_1 = \langle 1, 0, 0 \rangle a$ $a_2 = \langle 0, 1, 0 \rangle a$	$b_1 = \langle 1, 0, 0 \rangle \frac{2\pi}{a}$ $b_2 = \langle 0, 1, 0 \rangle \frac{2\pi}{a}$	$\langle 0, 0, 0 \rangle a$
Triangle Lattice	$a_1 = \langle 1, 0, 0 \rangle a$ $a_2 = \langle \frac{1}{2}, \frac{\sqrt{3}}{2}, 0 \rangle a$	$b_1 = \langle 1, -\frac{\sqrt{3}}{3}, 0 \rangle \frac{2\pi}{a}$ $b_2 = \langle 0, \frac{2\sqrt{3}}{3}, 0 \rangle \frac{2\pi}{a}$	$\langle 0, 0, 0 \rangle a$
Honeycomb Lattice	$a_1 = \langle 1, 0, 0 \rangle \sqrt{3}a$ $a_2 = \langle \frac{1}{2}, \frac{\sqrt{3}}{2}, 0 \rangle \sqrt{3}a$	$b_1 = \langle 1, -\frac{\sqrt{3}}{3}, 0 \rangle \frac{2\pi}{\sqrt{3}a}$ $b_2 = \langle 0, \frac{2\sqrt{3}}{3}, 0 \rangle \frac{2\pi}{\sqrt{3}a}$	$\langle \frac{\sqrt{3}}{4}, \frac{1}{4}, 0 \rangle a$ $\langle -\frac{\sqrt{3}}{4}, -\frac{1}{4}, 0 \rangle a$
Kagomé Lattice	$a_1 = \langle 1, 0, 0 \rangle 2a$ $a_2 = \langle \frac{1}{2}, \frac{\sqrt{3}}{2}, 0 \rangle 2a$	$b_1 = \langle 1, -\frac{\sqrt{3}}{3}, 0 \rangle \frac{2\pi}{2a}$ $b_2 = \langle 0, \frac{2\sqrt{3}}{3}, 0 \rangle \frac{2\pi}{2a}$	$\langle 0, \frac{\sqrt{3}}{3}, 0 \rangle a$ $\langle \frac{1}{2}, -\frac{\sqrt{3}}{6}, 0 \rangle a$ $\langle -\frac{1}{2}, -\frac{\sqrt{3}}{6}, 0 \rangle a$

5.1.4 Lasing From Holographic Dispersed Liquid Crystals Grating

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Starting from the pioneer works of Kogelnik and Shank [178] on distributed feedback (DFB) dye laser, the interests in compact organic laser device enhanced in recent years. For dye doped photonic crystal, the photonic crystal structure formed by H-PDLC acts as the optical cavity, while the doped dye molecules act as the laser working medium. As the dye molecules have a broad photoluminous spectrum, it is a good choice for tunable lasers. Lasing from 1D/2D H-PDLC PC has possible applications including: dynamic mirrorless all organic tunable laser device, “one step” easy fabricated organic laser, possible application in linear and nonlinear nano-photonics, and possible application as integer optical laser source.

As a good candidate of mirrorless organic devices, lasing from dye doped H-PDLC gratings attracted more and more concernings in research field. The reported H-PDLC-based laser emissions were mainly from dye-doped reflection or transmission gratings, where the underlying mechanism is much similar to conventional 1D distributed feedback lasers (DFB).

For 1D distributed feedback lasers, the active region of the device is periodically structured as a diffraction grating. The structure builds a one dimensional interference grating (Bragg scattering) and the grating provides optical feedback for the laser. DFB laser do not use two discrete mirrors to form the optical cavity (as they are used in conventional laser designs). The grating acts as the wavelength selective element for at least one of the mirrors and provides the feedback, reflecting light back into the cavity to form the resonator. The grating is constructed so as to reflect only a narrow band of wavelengths.

In 2003, R. Jakubiak et al [179] firstly reported the distributed feedback lasing action based on holographic polymer-dispersed liquid crystals switchable

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reflection grating with coumarin 485. G. S. He etc [180] reported tunable two-photon pumped lasing using a holographic polymer-dispersed liquid crystal reflection grating as a distributed feedback element. In their paper, the H-PDLC grating film was employed as an angle-dependent and narrow spectral-band feedback control element in the lasing generating system. In 2004, D. E. Lucchetta etc [181] realized lasing from a simple wafer structure made of a thin layer of rhodamine 6G as active material and of a switchable holographic mirror, basing on a reflection grating made of H-PDLC. In 2005, D. E. Lucchetta etc [182] investigated the fabrication technique and emission properties of novel all-organic laser devices based on reflection H-PDLC gratings with two different configurations: distributed Bragg-reflector configuration and distributed feedback configuration. As a lot of studies focused on properties of lasing from reflection H-PDCL grating [813-187], the study on lasing from H-PDLC transmission grating started from the report of V. K. S. Hsiao etc [188] in 2005, where an electrically switchable distributed-feedback lasing action was demonstrated in H-PDLC transmission grating structure. Comparing to reflection grating, the transmission grating provided a greatly enlarged gain length (~10 mm comparing to ~20 nm in common reflection grating) thus facilitated low threshold lasing with low pumping power. The low threshold, narrow line-width and liquid crystal concentration dependent property of lasing from H-PDLC transmission grating were also studied by Y. J. Liu in recent years [189-191].

5.2 Lasing From Two Dimensional Photonic Crystals

The lasing generated from two-dimensional photonic crystals can be divided into two types: one type is the lasing generated from bandgap defect-mode of photonic crystal with defect as cavity, where lasing action arise from the resonant cavity modes; the another type is the lasing generated from photonic crystal without defect, where no physically cavity exists and operation of laser is based on enhancement of the optical density of state due to small group velocity.

Within lasing produced from the second type of defect-free two dimensional photonic crystals, the mechanism differs due to the different refractive index contrast ratio of two materials consisting photonic crystals. For photonic crystals fabricated by relatively high refractive index contrast, e.g. semiconductor materials [192-194], and polymer [195-198], the lasing operates mostly at the band edge of photonic crystal band structure, which usually corresponds to high symmetry points, since at those points the group velocity is minimized thus maximized gain provided to oscillation.

For photonic crystals made by relatively low refractive index contrast materials, such as PbO glass [199] and H-PDLC, lasing action can be generated in a relatively wider range on corresponding band contrasting to certain points on band edge in the case of photonic crystal with relative large index contrast. In general, H-PDLC PCs cannot show complete photonic bandgaps due to the low refractive index contrast; however, local field enhancement is still possible through the group-velocity anomaly, where the group velocity becomes small over a wide range of wave vectors. The group-velocity anomaly, absent in one-dimensional (1D) case, is peculiar to 2D and 3D PCs and even existed in PCs with rather small index contrast.

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For H-PDLC materials, only 2D photonic crystals with square lattice has been demonstrated previously [200]. There is still quite large of room to study lasing from 2D photonic crystals H-PDLC. In this section, we will investigate the lasing generated from H-PDLC 2D PC with hexagonal lattice structure. Directional laser actions at specific wavelengths in two high-symmetry directions (ΓM and ΓK) are observed, while small group velocity due to group-velocity anomalies is responsible for the lasing.

5.3 Fabrication of Two Dimensional Holographic Polymer Dispersed Liquid Crystals Photonic Crystal

In our experiment, the LC/prepolymer mixture syrup used to fabricate the dye-doped 2D PC with hexagonal lattice structure was consisted of 53.10 wt% monomer, trimethylolpropane triacrylate (TMPTA), 13.14 wt% cross-linking monomer, *N*-vinylpyrrolidone (NVP), 0.72 wt% photoinitiator, Rose Bengal (RB), 1.09 wt% coinitiator, *N*-phenylglycine (NPG), 10.95 wt% surfactant, octanoic acid (OA), and 1.82 wt% lasing dye, 4-dicyanomethylene-2-methyl-6-*p*-dimethylaminostyryl-4H-pyran (DCM), all from Sigma-Aldrich, and 19.18 wt% liquid crystal, E7 ($n_o = 1.521$ and $n_e = 1.7462$), from Merck. (Add functions of each chemical component) The mixture was sandwiched in a cell, which was formed by two pieces of indium tin oxide (ITO) coated glass.

The 2D PC was holographically fabricated through the three-beam interference based on a single prism. Figure 5-6(a) shows the schematic of the prism used. The bottom plane (equilateral triangle) of the prism has a side length of $BC = BD = CD = 6$ cm, and a side-bottom plane angle of $\Phi = 60^\circ$. A collimated Ar⁺ laser beam (514.5 nm) impinged normally onto the prism, and

three beams, k_1 , k_2 , and k_3 (Figure 5-6 (b)) emerged through the refraction from three tilted side surfaces of the prism. In Figure 5-6(b), k represents wave vector, θ represents the angle between the wave vector and z axis, and φ represents the angle between the projection of wave vector on x - y plane and x axis. Here we have $\theta_1 = \theta_2 = \theta_3 = \theta = 25.3^\circ$ and $\varphi_1 = 0$, $\varphi_2 = 120^\circ$, $\varphi_3 = 240^\circ$. These three beams overlapped at the bottom surface of the prism and generated an interference pattern. The cell filled with LC/prepolymer mixture was attached onto the bottom surface using an index-matching liquid to record the interference pattern, as shown in Figure 5-6(c).

Figure 5-6 The specially designed prism, (b) three-beam interference configuration, and (c) schematic of the optical setup.

Assuming the three beams have the same phase, the intensity distribution of the interference pattern can be defined as [201]:

$$I(r) = \Re \left(\sum_{i,j=1}^3 E_i \cdot E_j \exp[i(k_i - k_j) \cdot r] \right), \quad (5.25)$$

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where E is the amplitude, $\mathbf{r} = (x, y, z)$ is a position vector, and $\Re[\dots]$ denotes the real part of the argument. A collimated plane wave was used to propagate normally onto the prism and be split into three beams. The wave vector of the three beams generated from the prism can be written as:

$$\mathbf{k}_m = k \left(-\cos \frac{2(m-1)\pi}{3} \sin \theta, -\sin \frac{2(m-1)\pi}{3} \sin \theta, -\cos \theta \right), \quad (5.26)$$

where m is from 1 to 3, $k = 2\pi n / \lambda$ is a unit wave vector, θ is the angle between the beam and vertical z axis, and λ is the writing wavelength in the air. Assuming the initial phases of three beams are equal to zero, we can obtain the interference pattern of hexagonal 2D photonic crystal in x - y plane.

$$\begin{aligned} I = & |E_1|^2 + |E_2|^2 + |E_3|^2 + 2E_1 \cdot E_2 \cos \left(\frac{3}{2} kx \sin \theta - \frac{\sqrt{3}}{2} ky \sin \theta \right) \\ & + 2E_1 \cdot E_3 \cos \left(\frac{3}{2} kx \sin \theta + \frac{\sqrt{3}}{2} ky \sin \theta \right) + 2E_2 \cdot E_3 \cos \left(\sqrt{3} ky \sin \theta \right). \end{aligned} \quad (5.27)$$

For three beams with equal intensity, $E_1 = E_2 = E_3$, the simulated interference pattern in x - y plane is shown in Figure 5-7(a), where the red- and blue-color areas represent high and low intensities, corresponding to polymer-rich and LC-rich regions respectively when it was recorded in the PDLCs. The lattice constant (shown in Figure 5-7(a)) in x - y plane is given by $a = 2\lambda / (3n \sin \theta)$. In our experiments, $\lambda = 514.5$ nm, $n = 1.537$ (calculated according to the ratio of polymer and LC), and $\theta = 25.3^\circ$, the theoretical lattice constant, a , is about 522 nm.

During the polymerization process, monomers start to photopolymerize in high intensity regions while LCs diffuse into low intensity region, thus forming the columnar polymer and LC droplets after the phase separation. The exposure intensity of each beam and exposure time were ~ 5 mW/cm² and 120 s,

respectively. The recording area of the 2D PC sample was about $5 \times 5 \text{ mm}^2$ in x - y plane with the thickness of $7 \text{ }\mu\text{m}$. Figure 5-7(b) shows the surface morphology of the 2D H-PDLC PC with a hexagonal lattice structure checked by an atomic force microscope (AFM), which was obtained after LC was removed in ethanol. The lattice constant was $\sim 500 \text{ nm}$ and the polymer columns (marked by the dash circle) have a radius of $\sim 145 \text{ nm}$. Considering a general 5% volume shrinkage for acrylate monomers during Photopolymerization [202], the experimental lattice constant (500 nm) was in good agreement with the prediction (522 nm). Figure 5-7(c) shows a scanning electron microscopy (SEM) image of the cross section morphology of the 2D H-PDLC PC, where the polymer columnar structure was observed along z direction. The morphology of the grating is slightly slanted due to a tilted SEM observation direction.

Figure 5-7 (a) Simulated intensity distribution of the three-beam interference pattern. (b) AFM image showing the surface morphology of the 2D hexagonal H-PDLC PCs. Scalar bar: 500 nm . (c) The cross section SEM image of H-PDLC sample. Scalar bar: $1 \text{ }\mu\text{m}$.

5.4 Generating Directional Lasing From Two Dimensional Holographic Dispersed Liquid Crystals Photonic Crystals

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Laser emissions along both ΓM (y axis) and ΓK (x axis) directions (Figure 5-7 (b)) in the sample took place when it was optically excited by a Q-switched frequency-doubled Nd:yttrium-aluminum-garnet (Nd:YAG) pulsed laser operating at 532 nm with a pulse duration of 7 ns and a repetition rate of 10 Hz. A linearly polarized pumping laser, focused by a cylinder lens, was incident on the surface of sample along the z direction with the transverse electric (TE) field in x - y plane. A fiber coupled spectrometer with a resolution of 0.6 nm was used to collect output lasing beams. The optical setup configuration is shown in Figure 5-8. The laser emission are expected to happen above 600 nm because there is an overlapping range in 500–590 nm for both absorption and photoluminescence (PL) spectra of DCM [190]. The absorption and PL spectra of DCM are shown in Figure 5-9 and Figure 5-10 respectively.

Figure 5-8 Schematic of laser generation optical configuration.

Figure 5-9 Absorption spectrum of DCM.

Figure 5-10 Photoluminescence (PL) spectrum of DCM.

5.5 Results and Discussion

Figure 5-11 shows the calculated photonic band structures for TE polarization in the 2D hexagonal HPDLC PC using the plane wave expansion method, where the parameters used were $r = 0.29a$, $n_p = 1.522$, and $n_{LC} = 1.597$.

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The laser emission occurred at 607 nm and 611 nm (Figure 5-12), corresponding to the normalized frequency of $\omega a/2\pi c = 0.824$ and 0.818 along ΓM and ΓK direction respectively (Figure 5-11). From Figure 5-11, we can see that the bands, where the lasing happened along ΓM and ΓK directions respectively, are relatively flat, indicating the small group velocities. In a gain medium, a small group velocity v_g causes a long effective gain length [203], where the gain is inversely proportional to v_g , and thus leads to a large enhancement of local fields—light amplification [200]. Therefore, the laser emission was induced by the group-velocity anomalies in flat photonic bands.

Figure 5-11 Photonic band structure of the 2D hexagonal H-PDLC PC for the TE polarization. The inset represents the irreducible Brillouin zone. Arrows show the located frequencies where the lasing action occurs.

Figure 5-12(a) shows the lasing spectra measured along ΓM direction under different pumping energies. The lasing peak centers at 607 nm with a linewidth of 1.5 nm. The peak intensity as a function of pumping energy is illustrated in Figure 5-12(b). The threshold of 180 $\mu J/pulse$ is achieved. The same measurements along ΓK direction were carried out, as shown in Figure 5-

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12(c) and 5-12(d). There were no significant polarization characteristics in both ΓM and ΓK directions. Compared with ΓM case, a slightly redshifted lasing peak occurs at 611 nm with the linewidth of 1.6nm (Figure 5-12(c)), and a much lower threshold of about 70 $\mu J/pulse$ is obtained (Figure 5-12(d)). The laser threshold is proportionally related to the group velocity [204]. From Figure 5-11, the band along ΓK direction is much flatter than that along ΓM direction, which induces a smaller group velocity but a larger enhancement of the local fields, thereby leading to a lower threshold. It is also worth mentioning that besides the primary peak, several other competing peaks appeared in the spectra along both ΓM and ΓK directions due to the spatial hole burning effect [205] when the pumping energy was increased far beyond the threshold (red curves in Figure 5-12(a) and 5-12(c)).

Figure 5-12 Lasing spectra measured along (a) ΓM and (c) ΓK directions. Relationships of peak intensity verse pumping energy along (b) ΓM and (d) ΓK directions.

5.6 Conclusion

In conclusion, we demonstrated lasing actions along both ΓM and ΓK directions from a DCM dye-doped 2D hexagonal H-PDLC PC. The 2D photonic crystal with hexagonal lattice structure was fabricated by three beam holography interference. The experimental results of photonic crystal structure were consistent with simulation result. The photonic crystal band structure,

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which was analysed by plane wave expansion method, was used here to help understanding the underlying mechanism of lasing oscillations. The group velocity anomaly was the underlying mechanism for the substantial light amplification in our 2D H-PDLC PCs with the low-index contrast. Lasing spectra under different pumping power level was measure as well as the laser threshold characteristics in both directions. The big difference on the laser threshold along ΓM and ΓK directions was explained through the proportional relationship of the laser oscillation threshold and the group velocity of the corresponding band.

Chapter 6 Electric Field and Temperature Effects on Lasing from Two-Dimensional Holographic Polymer Dispersed Liquid Crystal Photonic Crystal with Hexagonal Lattice Structure

6.1 Introduction

In 2D H-PDLC photonic crystals, the orientation of LC droplets is assumed to be randomly or partly formed by the two-dimensional matrix structure of polymers depending on specific fabrication environmental parameters such as intensity, exposure time, concentration ratio of LC and monomer, and structures formed. The orientation of LC droplets directors in polymer matrix can be influenced by many factors such external electric field, and temperature. Under applied voltage, the directors of liquid crystal droplets formed in H-PDLC will reorient according to the electric field built in, transferring the status of randomly distributed LC droplets with isotropic index or small anisotropic index to a status of regularly aligned LC droplets with anisotropic index. Beside electric field, the index of LC is also dependent on temperature. When the environmental temperature increases, the ordinary and extraordinary refractive index of LC will increase and decrease in the range of liquid crystal phase, respectively. Once the environmental temperature increases beyond the clear temperature of liquid crystal, liquid crystal phase will be changed to an isotropic liquid phase.

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The tunable property of lasing based on dye-doped H-PDLC has been discussed such as in reflection grating [181, 182], transmission grating [184, 188], and 2D square PC [201] in the presence of electric field. The electrical erasing and shifting of lasing in H-PDLC has also been discussed. However, the polarization characteristics of lasing under applied voltage have not been investigated yet, especially in 2D PC. Under applied voltage, the refractive index contrasts of 2D PC for the two perpendicular polarizations change differently with respect to each other because of the realignment of the nematic directors in the LC droplets along the electric field, which possibly leads to different lasing spectra corresponding to transverse electric (TE) and transverse magnetic (TM) modes. The temperature dependent tunable properties of lasing have also been studied in PDLC [206] and H-PDLC transmission grating [191], however, rarely in 2D H-PDLC PCs.

In this section, we will demonstrate the electrically tunable property of lasing for TE and TM modes in a dye-doped 2D H-PDLC PC with hexagonal lattice. The splitting of laser spectra between TE and TM modes is observed and subsequently explained by the opposite band shifting of TE and TM modes. Also, we systematically study the temperature dependence of lasing from 2D H-PDLC PC with hexagonal lattice structure along Γ M direction in TE polarization. We report a red-shift of lasing peaks with the increase of temperature, which can be explained by the downward movement of photonic band, which is primarily due to the decrease of LC droplets' anisotropy with the increasing ambient temperature.

6.2 Electric Field effect on Lasing from Two-Dimensional

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6.2.1 Experiment

In our experiment, the LC/prepolymer mixture syrup that is used to fabricate the dye-doped 2D PC with hexagonal lattice structure consisted of 63.76 wt% monomer, trimethylolpropane triacrylate (TMPTA), 7.05 wt% cross-linking monomer, N-vinylpyrrolidone (NVP), 0.49 wt% photoinitiator, Rose Bengal (RB), 0.98 wt% coinitiator, *N*-phenylglycine (NPG), 9.30 wt% surfactant, octanoic acid (OA), and 1.18 wt% lasing dye, 4-dicyanomethylene-2-methyl-6-p-dimethylaminostyryl-4H-pyran (DCM), all from Sigma-Aldrich, and 17.24 wt% liquid crystal, E7 ($n_o = 1.5216$ and $n_e = 1.7462$), from Merck. The mixture was sandwiched in a cell, which was formed by two pieces of indium tin oxide (ITO) coated glass. The cell gap $d = 7 \mu\text{m}$.

The 2D H-PDLC PC with hexagonal lattice structure was holographically fabricated through the three-beam interference based on a single prism using collimated Ar⁺ laser beam (514.5 nm). The exposure intensity of each beam was 5 mW/cm^2 , with an exposure time of 120 s. The recording area of the fabricated 2D PC sample was about $5 \times 5 \text{ mm}^2$ in x - y plane. During the polymerization process, monomers start to photopolymerize in high intensity regions while LCs diffuse into low intensity region, thus forming the columnar polymer and LC droplets after the phase separation.

Figure 6- 1(a) AFM image showing the surface morphology of a 2D hexagonal H-PDLC PC, a is the lattice constant. Scale bar: 1 μm . (b) Schematic optical setup of the lasing experiment.

Figure 6-1(a) shows the surface morphology of the 2D H-PDLC PC with a hexagonal lattice structure imaged by an atomic force microscope (AFM). The lattice constant of the 2D PC sample, a , was ~ 515 nm and the polymer columns have a radius of ~ 152 nm. The lattice constant in x - y plane can be calculated by $a = 2\lambda / (3n\sin\theta)$. In our experiments, $\lambda = 514.5$ nm, $n = 1.535$ (calculated according to the ratio of polymer and LC), and $\theta = 25.3^\circ$, the theoretical lattice constant is about 523 nm. The detail fabrication experiment can be found in chapter 5. Considering a general 5% volume shrinkage for acrylate monomers during photopolymerization, the experimental lattice constant (515 nm) was in good agreement with our prediction (523 nm).

The schematic optical setup of our lasing experiment is shown in Figure 6-1(b). To generate lasing from the 2D H-PDLC sample, a Q-switched frequency-doubled Nd:yttrium-aluminum-garnet (Nd:YAG) pulsed laser operating at 532 nm, with a pulse duration of 7 ns and a repetition rate of 10 Hz, was used. A linearly polarized (electric field in y direction) pumping laser,

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focused by a cylinder lens, was incident on the surface of the sample along the z direction. A fiber coupled spectrometer positioned along ΓM direction with a resolution of 0.6 nm was used to collect output lasing beams. The experiment was performed at room temperature (25°C).

6.2.2 Results and Discussions

Figure 6-2 shows the changes of transmission and diffraction efficiency of our 2D H-PDLC PC sample (with incident angle $\sim 40^\circ$, where the incident angle is defined as the angle between the probe laser propagation direction and the normal line of glass-substrate-plane) as functions of applied voltage. A linear polarized white light (polarization direction along y axis) was used as illuminate source. Without applied voltage, the diffraction and transmission of our 2D H-PDLC PC showed no change when rotate the polarization of incident white light in x - y plane, which indicated the initial state of LC droplet are isotropic. As the applied voltage increased, the transmission and diffraction efficiency increased and decreased respectively. There was still no significant difference of transmission and diffraction efficiency among incident lights with different polarizations in x - y plane. The threshold behavior was observed at 15 V.

Figure 6-2 Changes of transmission and diffraction efficiency of our 2D H-PDLC PC sample as functions of applied voltage.

Figure 6-3(a)-(c) show the lasing spectra from ΓM direction of our 2D PC sample at the externally applied voltages of 0 V, 25 V, and 40 V respectively, all with the fixed optical pumping energy of 380 $\mu J/pulse$. The green curves represent the lasing spectra obtained without polarizer, and their corresponding lasing peaks are 613/617 nm, 622/627 nm and 625/631 nm under 0 V, 25 V and 40 V, respectively. Those lasing peaks were due to the group-velocity anomalies, which are peculiar to 2D and 3D PCs. The simulation works reported by Sakoda [207] indicated that two or more lasing peaks can be generated simultaneously in the frequency range of group-velocity anomalies of 2D PC with a small index contrast, which gives a good explanation of our experimental spectra with multi-lasing peaks here. Additionally, these competing lasing peaks appear at high pumping energy levels, which could be due to the spatial hole burning effect. In our experiment, over 10 nm red-shifting of the non-polarized lasing wavelength with the increased voltage was obtained.

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Figure 6-3 Lasing spectra measured along ΓM directions under the applied voltage of (a) 0 V, (b) 25 V, and (c) 40 V. Green, red and blue curves represent the spectra of non-polarized, TE mode and TM mode lasing, respectively. Red and blue dash lines represent the central lasing peaks for TE and TM modes under applied voltage, respectively.

The directors of the liquid crystal droplets in H-PDLC will be aligned by external electric field, which produces a larger difference in the photonic crystal band structure of TE and TM modes and finally leads to different laser spectra for them. In Figure 6-3, the laser spectra of TE and TM modes are represented by red and blue curves, respectively. From Figure 6-3(a), we can see that the lasing spectra show no significant difference for TE and TM modes, as the peaks of 613/617 nm in non-polarized spectra are reserved both in TE and TM case. When the external voltage applied exceeded the threshold of 15 V (as shown in Figure 6-3(b) and 6-3(c)), two phenomena are observed: First, the lasing splitting spectra of TE and TM modes started to appear and enlarged as

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the voltage increased; Second, the lasing spectrum of TE and TM modes red-shifted as the voltage increased. In the following sections, we are trying to find out the underlying reasons for these two interesting phenomena.

To further investigate the lasing in dye-doped 2D H-PDLC PC, the photonic band structures were calculated for the 2D hexagonal HPDLC PC using the plane wave expansion method, shown in Figure 6-4(a). Here the band structures of TE and TM modes are represented by red and blue solid line, respectively, where $r = 0.30a$, $n_p = 1.522$, and $n_{LC} = 1.597$. The tunable lasing described in Figure 6-3 ranges from 613 to 637 nm, corresponding to the normalized frequency range from $\omega a / 2\pi c = 0.808$ to 0.840 along ΓM direction. The band where laser actions occurred is marked by the arrow. It is noticed that the index contrast is quite small in our 2D PC sample, therefore leading to the strong overlap of the TE and TM band structures (as shown in Figure 6-4(a)), which leads to the non-split lasing spectra of TE and TM modes when no voltage is applied.

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Figure 6-4 Photonic band structure of the 2D hexagonal H-PDLC PC. The inset represents the irreducible Brillouin zone. Red and blue solid line represent the TE and TM modes, respectively, where $r = 0.30a$, $n_p = 1.522$, and $n_{LC} = 1.597$. (b) The shift of the band where the laser action occurred under at the applied voltage. Red dash line represents TE mode and blue dash line represents TM mode. The arrows represent the shifting directions with increasing voltage. A, B, and C represent possible lasing points on photonic band, respectively.

However, when an external voltage was applied, the effective index of liquid crystal droplets started to decrease for TE mode and increase for TM mode, leading to the index contrast of LC droplets and polymer decrease and increase for TE and TM modes, respectively. Figure 6-4(b) shows the shifting trend of the band with increasing external applied voltage. The decreasing and increasing of effective refractive index for TE and TM modes yields the upward shifting (towards shorter wavelengths) for the band of TE mode (red dash line) and downward shifting (towards longer wavelengths) for the band of TM mode (blue dash line), which in turn leads to the relatively shorter wavelength of lasing peaks in TE mode and longer wavelength of lasing peaks in TM mode, as shown in Figure 6-3(b) and 6-3(c). The shift directions of TE and TM bands are shown by the arrows. Therefore, in the presence of an external voltage, the

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directors of the liquid crystal droplets in H-PDLC will be aligned, which produces the larger difference of photonic crystal band structure for TE and TM mode, from opposite band shift directions, and finally leading to the different lasing spectra for TE and TM modes.

If applied voltage further increases, the band difference of TE and TM modes will become bigger (not shown here), due to the further decreased and increased effective index of LC for TE and TM mode respectively. It means the splitting spacing of peaks for TE and TM modes will increase with the increased electric field. From Figure 6-3(a), we can see that at voltage of 0 V, the laser peaks of TE and TM modes are overlapped, no splitting spacing occurred here; In Figure 6-3(b), at voltage of 25 V, the spacing of peaks for TE and TM modes is $627\text{ nm}-622\text{ nm}=5\text{ nm}$ (distance of dash red and blue lines); In Figure 6-3(c), at voltage of 40 V, the spacing of peaks could be considered as the difference of central peaks for TE and TM modes (distance of dash red and blue lines): $631\text{ nm}-620\text{ nm}=11\text{ nm}$. Therefore, the splitting spacing of the peaks for the TE and TM modes is increased as the increasing electric field, which is quite consistent with the prediction from photonic band.

For the red-shift of lasing spectrum both for TE and TM modes as the voltage increased in our experiment, the possible reason is as follows: because of the property of lasing induced from group velocity anomaly, the possible range of lasing frequencies is within the photonic band, which means that the generated laser peaks could be different under different specific oscillation conditions (e. g. different voltages). Assuming the oscillation is initially at P1 position (with lasing point A on the band) without voltage (shown in Figure 6-4(b)). When voltage is applied, the condition changes corresponding and the

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oscillation position might be shifted to P2, accompanied by the split of TE and TM band (with lasing point B on TE and point C on TM band). Therefore, the movement from P1 to P2 and the split of TE and TM with voltage applied lead to two results: 1) Comparing to point A, the laser wavelengths on points B and C are larger in absolute value, meaning a red-shift of lasing spectrum; 2) The split of points B and C leads to the laser splitting for TE and TM modes. Even the reason of why the laser oscillation position was red-shifted (towards to Γ) but not blue-shifted (towards to M) with the increased voltage in our experiment is still unclear, but the fact of laser wavelength shifting within the range of photonic band made the experimental results still reasonable and understandable.

6.3 Temperature Dependent Property of Lasing from Two-Dimensional Holographic Polymer Dispersed Liquid Crystals Photonic Crystal with Hexagonal Lattice Structure

6.3.1 Experiment

In our experiment, the 2D H-PDLC PC with hexagonal lattice structure was holographically fabricated through the three-beam interference based on a single prism impinging by collimated Ar⁺ laser beam (514.5 nm). During the polymerization process, monomers start to photopolymerize in high intensity regions while LCs diffuse into low intensity region, thus forming the columnar polymer and LC droplets after the phase separation. The exposure intensity of each beam was 15 mW/cm², with an exposure time of 120 s. The recording area of the fabricated 2D PC sample was about 5×5 mm² in *x-y* plane.

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The LC/prepolymer mixture syrup used to fabricate the dye-doped 2D PC with hexagonal lattice structure consisted of 63.76 wt% monomer, trimethylolpropane triacrylate (TMPTA), 7.05 wt% cross-linking monomer, *N*-vinylpyrrolidone (NVP), 0.49 wt% photoinitiator, Rose Bengal (RB), 0.98 wt% coinitiator, *N*-phenylglycine (NPG), 9.30 wt% surfactant, octanoic acid (OA), and 1.18 wt% lasing dye, 4-dicyanomethylene-2-methyl-6-p-dimethylaminostyryl-4H-pyran (DCM), all from Sigma-Aldrich, and 17.24 wt% liquid crystal, E7 ($n_o = 1.5216$ and $n_e = 1.7462$), from Merck. The mixture was sandwiched in a cell, which was formed by two pieces of indium tin oxide (ITO) coated glass. The cell gap was 7 μm .

Figure 6-5 Schematic optical setup of the lasing experiment. Inset figure is an AFM image for the surface morphology of a 2D hexagonal H-PDLC PC. Scale bar: 500 nm.

To generate lasing from the 2D H-PDLC sample, a Q-switched frequency-doubled Nd:yttrium-aluminum-garnet (Nd:YAG) pulsed laser operating at 532 nm, with a pulse duration of 7 ns and a repetition rate of 10 Hz,

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was used. A linearly polarized pumping laser, focused by a cylinder lens, was impinged on the surface of the sample along the z direction in TE polarization (electric field in x - y plane). A fiber coupled spectrometer along x axis with a resolution of 0.6 nm was used to collect output lasing beams, and the output laser was measured in TE polarization (with polarization along y axis). The schematic optical setup is shown in Figure 6-5, and the inset figure shows the surface morphology of the 2D H-PDLC PC with a hexagonal lattice structure imaged by an atomic force microscope (AFM). The lattice constant of the 2D PC sample, a , was ~ 517 nm, and the polymer columns have a radius r of ~ 155 nm.

6.3.2 Results and Discussions

Figures 6-6(a)-(c) show the lasing spectra measured along ΓM direction at temperatures of 25, 35 and 45°C, under different pumping energies of 260, 300, and 380 $\mu\text{J/pulse}$. The corresponding lasing peaks, which increase with the increasing pumping energy, are 610/615 nm, 620/626 nm, and 624/631 nm in Figure 6-6(a)-(c) respectively. Those lasing peaks were induced by the group-velocity anomalies, which is peculiar to 2D and 3D PCs and even exists in PCs based on H-PDLC materials with rather small index contrast. The simulation works reported by Sakoda indicated that two or more lasing peaks can be generated simultaneously in the frequency range of group-velocity anomalies of 2D PC with small index contrast [207], which gives a good explanation of our experimental spectra with two lasing peaks here. Additionally, although allowed in theory, these completing lasing peaks are preferred to appear under a higher pumping energy, due to the spatial hole burning effect. From Figure 6-

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6(a)-(c), we can find that when the temperature is increased, the intensity of lasing peaks is decreased and the wavelength of lasing peaks is red-shifted.

Figure 6-6 Lasing spectra measured along ΓM direction at temperatures of (a) 25°C, (b) 35°C, and (c) 45°C, under different pumping energies of 260 $\mu J/pulse$, 300 $\mu J/pulse$, and 380 $\mu J/pulse$.

Figure 6-7(a) shows the dependence of the lasing peak intensity (610 nm) and the full width at half maximum (FWHM) of the peak as functions of pumping energy at 25°C. The pumping threshold energy (P_{th}) was around 200 $\mu J/pulse$, above which the peak intensity and FWHM increased and narrowed rapidly respectively with the increase of pumping energy. A 1.9 nm FWHM of lasing peak was obtained at 25°C, under the pumping energy of 380 $\mu J/pulse$ here. It is worth noticing that, under the same pumping energy of 380 $\mu J/pulse$, the FWHM of lasing peak at different temperatures, e. g. 620 nm for 35°C and 624 nm for 45°C, showed little change compared to that at 25°C. The dependence of lasing peak intensity on temperature from 25°C to 60°C is shown in Figure 6-7(b), where the pumping energy is fixed at 380 $\mu J/pulse$. As the

temperature increases, the lasing peak intensity decreases, showing a similar behavior to the previous report on lasing from dye-doped H-PDLC transmission grating [191]. With the increase of temperature, the average effective index of LC will decrease, leading to a reduced index difference of LC droplets and polymer [164], thus a weaker light scattering of H-PDLC sample to input light. Therefore, the output light intensity was reduced accordingly.

Figure 6-7 (a) Dependences of peak intensity and FWHM of PL maximum peak on pumping energy at 25°C. The threshold of pumping energy (P_{th}) is around 200 $\mu\text{J}/\text{pulse}$. (b) Dependence of lasing peak intensity on temperature. The pumping energy is 380 $\mu\text{J}/\text{pulse}$.

In general, the H-PDLC 2D PC is slightly anisotropic since the long narrow morphology of the LC droplets tends to align the liquid crystals parallel to the z direction [208]. Therefore the index of LC droplets experienced by TE polarization (n_{LC}) at temperature of 25°C is smaller than the average refractive index of LC ($n_{ave}=(2n_o+n_e)/3$). When the temperature is increased, two mechanisms would influence the refractive index of LC droplets experienced by the TE polarized light. Firstly, the optical anisotropy will be reduced and the TE

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polarization will experience a higher index of LC droplets, approaching the average refractive index of LC. The actual index change relies on the initial optical anisotropy of the LC droplets (the largest possible change is up to $\Delta n = n_{ave} - n_o \approx 0.07$). Secondly, the n_{ave} of LC will be decreased with the increase of temperature, but this effect is comparably small ($\Delta n_{ave} \approx -0.01$ from 25 to 45°C) [164]. The final LC index experienced by the TE polarization will depend on these two competing mechanisms. As we discussed above, the optical anisotropy reduction should be the dominant factor. In our experiment, the red-shift of lasing spectrum (corresponding to an increased refractive index) with the increase of temperature indicates that indeed the influence from the reduced optical anisotropy is dominant.

Figure 6-8 (a) Photonic band structure of the 2D hexagonal H-PDLC PC for TE polarization, where $r = 0.30a$, $n_p = 1.522$, and $n_{LC} = 1.570$. (b) The shift of the band where the laser action occurred with the increase of temperature. Red solid line and green dash line represent the band of TE polarization with $n_{LC} = 1.570$ and with $n_{LC} = 1.597$, respectively. The arrows represent the shifting directions with increasing temperature.

The photonic band structure of our 2D hexagonal H-PDLC PC, calculated by the plane wave expansion method, is shown in Figure 6-8 (a), assuming $r = 0.30a$, $n_p = 1.522$, and the initial index of LC for TE polarization at the temperature of 25°C, $n_{LC} = 1.570$. The solid red line represents the photonic band of TE polarization and the band where the generated laser is marked by the arrow. When temperature is increased, assuming the experienced index of LC droplets by TE polarization increased from $n_{LC} = 1.570$ to 1.597 and the index of polymer is unchanged, the photonic band of TE polarization shifted downward, as shown in Figure 6-8(b) by the green dash line. The downward shift of photonic band, shown by the arrow, leads to a decrease in the lasing frequency, therefore an increase in wavelength. Therefore, the red-shift of the experimental lasing spectra with increased temperature (in Figure 6-6), is quite consistent with our theoretical analysis discussed above.

6.4 Conclusion

We demonstrated the electrically tunable properties of lasing with multi-peaks from a dye-doped 2D H-PDLC PC with hexagonal lattice. Over 10 nm red-shifting of the lasing peaks were obtained in non-polarization spectra when the applied voltage was increased up to 40 V. The effect of the applied electric field on the effective refractive index of LC are, however, also different for the TE and TM modes, which lead to an opposite band shifting and eventually result in the splitting of the lasing spectra between TE and TM modes. We also demonstrated the temperature tunable properties of lasing from a dye-doped 2D H-PDLC PC with hexagonal lattice along ΓM direction. When the temperature increased from 25 to 45°C, a red-shift of lasing peaks has been observed. The

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decrease of LC droplets' anisotropy with the increased temperature for TE polarization is the major underlying reason for downward movement of photonic band, which eventually leads to the red-shift of lasing peaks.

Chapter 7 Conclusion and Future Works

7.1 Conclusion

In this dissertation, the studies on properties of different new liquid crystals based diffractive optical elements and investigations on lasing from two-dimensional holographic polymer dispersed liquid crystals photonic crystals devices were presented.

For liquid crystals based diffractive optical elements parts. We firstly demonstrated an electrically-switchable CGH using LC cell with proton beam patterned PMMA film. The CGH desired for generating words of NTU was firstly generated by computer and then fabricated on PMMA substrate coating on ITO glass by proton beam writing method. Comparing to the CGH recorded in PDLC where a transparency mask was used with a resolution of $50\mu\text{m}$, a higher resolution ($\sim 2\mu\text{m}$) was achieved by proton beam, leading to a better quality of the reconstructed CGH image. It has less light scattering and relatively lower operating voltage ($\sim 15\text{ V}_{\text{rms}}$), compared to the CGH using a PDLC material. Higher cost and longer fabrication time are the drawbacks of this method. The maximum diffraction efficiency measured is about 28.7%. This device is promising in adaptive optical elements.

Secondly, a pair of electrically switchable 2D finite energy Airy beams has been generated by a liquid crystal cell with binary-phase patterned electrode. These Airy beams are electrically switchable, which can be generated or erased with the change of applied electric field. The diffraction-free and transverse acceleration dynamics of the Airy beams were verified. This kind of binary-phase element based Airy beam device has the advantages of low cost,

easy fabrication process, and electrical tunability, potentially useful in adaptive optics.

Finally, we have generated a polarization-dependent circular Dammann grating diffraction from an azo-dye-doped LC cell. A simple multi-exposure photo-aligned process was used to fabricate a binary phase liquid crystal CDG zone plate that consisted of an even zone with homogenous LC structure and an odd zone with 60° and 55° TN LC structure. The maximum 0^{th} and $1^{\text{st}/2^{\text{nd}}}$ diffraction orders of CDG can be separately achieved by changing the analyzer's polarization direction. If the chosen analyzer's direction leads to a zero phase difference in output light from even and odd zones, the maximum 0^{th} diffraction order will be achieved. In contrast, if the chosen analyzer's direction leads to a π phase difference in output light from even and odd zones, the maximum 1^{st} diffraction order will be produced. The CDG diffraction from our azo-dye-doped LC cell can be generated by lasers within a broad wavelength band, which expands possible applications.

For the lasing from two-dimensional holographic polymer dispersed liquid crystals photonic crystals devices parts. We firstly demonstrated lasing actions along both ΓM and ΓK directions from a DCM dye-doped 2D hexagonal H-PDLC PC. The 2D photonic crystal with hexagonal lattice structure was fabricated by three beam holography interference. The experimental results of photonic crystal structure were consistent with simulation result. The photonic crystal band structure, which was analysed by plane wave expansion method, was used here to help understand the underlying mechanism of lasing oscillations. The group velocity anomaly was the underlying mechanism for the substantial light amplification in our 2D H-

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PDLC PCs with the low-index contrast. Lasing spectra under different pumping power level was measure as well as the laser threshold characteristics in both directions. The big difference on the laser threshold along ΓM and ΓK directions was explained through the proportional relationship of the laser oscillation threshold and the group velocity of the corresponding band.

Then, we demonstrated the electrically tunable properties of lasing with multi-peaks from a dye-doped 2D H-PDLC PC with hexagonal lattice. Over 10 nm red-shifting of the lasing peaks were obtained in non-polarization spectra when the applied voltage was increased up to 40 V. The effect of the applied electric field on the effective refractive index of LC are, however, also different for the TE and TM modes, which lead to an opposite band shifting and eventually resulted in the splitting of the lasing spectra between TE and TM modes. We also demonstrated the temperature tunable properties of lasing from a dye-doped 2D H-PDLC PC with hexagonal lattice along ΓM direction. When the temperature increased from 25 to 45°C, a red-shift of lasing peaks has been observed. The decrease of LC droplets' anisotropy with the increased temperature for TE polarization is the major underlying reason for downward movement of photonic band, which eventually leads to the red-shift of lasing peaks.

7.2 Future Works

In this dissertation, tunable diffractive optical elements were realized based on nematic liquid crystals and its special characteristics. For diffractive optical elements, the tunable properties can be improved and extended by using other kinds of liquid crystal such as cholesteric LC, ferroelectric LC, and blue phase LC etc while more novel tunable diffractive optical elements can be

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realized through nematic liquid crystals materials. Also, as all diffractive optical elements discussed here are binary-phase elements, although the binary-phase elements have advantages of easy fabrication and simple simulation and calculation process, the low efficiency is a major drawback for such kind of diffractive optical elements. To solve the intrinsic drawback of binary-phase, more efforts are needed on developing and realizing liquid crystal tunable diffractive optical elements based on multi-level phase comparing to binary phase.

On the other side, with realization of quasi photonic crystal using H-PDLC becomes possible by five, seven, or twelve beams interference, the researches on lasing from 2D quasi H-PDLC photonic crystal devices would be a very interesting field. As previously reported results are majorly focused on lasing from quasi photonic crystal fabricated by semiconductor materials with relatively high index contrast, the research on lasing from H-PDLC quasi photonic crystals could help complete and explain the mechanism behind the lasing from quasi photonic crystal based on all organic materials with low index contrast. Besides, the research on lasing from 2D H-PDLC can be extend to 3D case, as 3D photonic crystal could provide more free degrees of lasing directions as well as more possible oscillate wavelengths. The electrically and temperature dependent properties of nematic liquid crystals also help to realize tunable lasing from such quasi and 3D H-PDLC photonic crystals.

Author's Publications

Journal Paper

1. **D. Luo**, H. T. Dai, X. W. Sun, and H. V. Demir, "Polarization-dependent circular Dammann grating made of azo-dye-doped liquid crystals," *Appl. Opt.* **50**, 2316-2321 (2011).
2. **D. Luo**, H. T. Dai, X. W. Sun, and H. V. Demir, "Electrically switchable finite energy Airy beams generated by a liquid crystal cell with patterned electrode," *Opt. Commun.* **283**, 3846-3849 (2010).
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Conference Paper

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