REFLECTION AND TRANSMISSION OF OBLIQUELY INCIDENT LIGHT BY ASYMMETRIC SERIAL-BIDEPOSITED CHIRAL SCULPTURED THIN FILMS

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by

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Abstract

A chiral sculptured thin film (STF) exhibits the Circular Bragg Phenomenon (CBP), which is the differential reflection of incident light of left- and right-circular polarization states in a spectral regime called the circular Bragg regime. One way to fabricate a chiral STF is to direct a collimated vapor flux obliquely towards a uniformly rotating substrate mounted inside a vacuum chamber, the fixed angle between the vapor flux and the substrate plane being denoted by $\chi$. This process is often implemented by rotating the substrate by an angle $\delta \ll \pi$ and a subdeposit of height $h$ is deposited before the next rotation by $\delta$, resulting in a finely ambichiral STF. Another way is the asymmetric serial-bideposition (ASBD) technique, whereby the substrate is rotated alternately by angles $\pi$ and $\pi + \delta$, $\delta \ll \pi$, and subdeposits of heights $h_1$ and $h_2 = h_{x:1} - h_1$ are sequentially deposited with $h_{x:1} = h_1 + h_2$ fixed.

I investigated the effect on the CBP by altering the ratio $h_1:h_2$ while keeping $h_{x:1}$ fixed. Structurally right-handed chiral STFs of zinc selenide were deposited with ratios 1:1, 1.5:1, 2:1, 2.5:1, 3:1, 5:1, 7:1, and 9:1, while $h_{x:1} = 2.17$ nm, $\chi = 20^\circ$, and $\delta = 3^\circ$ were kept fixed. A finely ambichiral STF was also deposited with $h = 2.49$ nm, $\chi = 20^\circ$, and $\delta = 3^\circ$. The period of all samples was fixed at 300 nm and the number of periods at 10. Measurements of the circular reflectances ($R_{LL}$, $R_{LR}$, $R_{RL}$, and $R_{RR}$) were made with the angle of incidence $\theta_{inc}$ varying between $10^\circ$ to $70^\circ$, while measurements of the circular transmittances ($T_{LL}$, $T_{LR}$, $T_{RL}$, and $T_{RR}$) were made for $\theta_{inc} \in [0^\circ, 70^\circ]$. Red-shifting and narrowing of the circular Bragg regime were found to intensify with increasing $h_1:h_2$ for all values of $\theta_{inc}$. A limit in red-shifting seemed to be achieved with the 9:1 sample, with a better defined circular Bragg regime than that of the finely ambichiral sample.
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Chapter 1

Introduction

1.1 Overview

The morphology of a sculptured thin film (STF) is engineered by manipulating the growth direction of parallel nanocolumns during oblique-angle physical vapor deposition (PVD). Chiral STFs are assemblies of parallel helical nanocolumns separated by a void network, and have been experimentally and theoretically proven to exhibit the Circular Bragg Phenomenon (CBP), whereby a chiral STF of a certain structural handedness will highly reflect circular-polarized light of the same handedness, but not circular-polarized light of the opposite handedness [1], [2]. The CBP is displayed in a spectral regime called the circular Bragg regime. Since the conceptualization of chiral STFs, most experimental research has been focused on light incident normally on a chiral STF so that the propagation vector of incident light is parallel to the common axis of the helical nanocolumns. If the angle
of incidence $\theta_{inc}$ between the propagation vector of the incident light and the common axis of the helical nanocolumns is increased, the central wavelength $\lambda_{Br}^0$ of the circular Bragg regime blue shifts. This has been predicted theoretically [3], and demonstrated experimentally for zinc selenide in the visible regime [4].

Besides optical applications, chiral STFs have biomedical, electronic, and mechanical applications. Theoretical studies of chiral STFs confined between two metallic planes were undertaken by Ertekin and Lakhtakia [5]. Functioning as an optical interconnect, this structure could help increase optoelectronic circuit board chip-density [5]. The advancement of optical interconnects along with multiplexing is guaranteed to send computers into the exa-scale (a billion billion computations per second) age [6].

Seto et al. were able to show that helical nanocolumns of a chiral STF (with a capping layer to distribute the force) respond elastically to nano-indentations, and were able to extract a spring constant and resonance frequency of the chiral STF [7].

The inter-nanocolumnar void network produced during oblique-angle PVD provides space for infiltration with chemical and biological substances for sensing applications by altering the effective constitutive properties of the chiral STF. Lakhtakia conceptualized a bilayer consisting of two chiral STFs of opposite handedness to sense toxic gases [8]. A clear shift in the bandwidth $\Delta \lambda_{Br}^0$ and the central wavelength $\lambda_{Br}^0$ of the circular Bragg regime was calculated when going from 0 to
50 mole/m³ of adsorbed gas [8]. Since then, sensing of various fluids has been experimentally verified [9–12].

This chapter goes through the history of how STFs in general arose from more simple variations of columnar thin films (CTFs), how such morphologies can be engineered via PVD, and the motivation for the original research reported in this thesis. The CBP exhibited by chiral STFs is discussed in Chapter 2. Chapter 3 presents the experimental methods used for growing all the samples and for their optical and morphological characterization. The optical responses along with measures of the quality of the circular Bragg regimes are presented in Chapter 4. Conclusions from the obtained results are provided in Chapter 5 as well as ideas for future work.

1.2 Timeline of STFs

Long before the advent of STFs, it was realized that the simpler CTFs could be produced by controlling certain parameters during film growth as well as by holding the substrate fixed at an angle with respect to the direction of a collimated vapor flux of the material to be deposited [13]. However, before any such films could be realized, the methods of fabricating them first needed to exist, and hence these methods are now discussed.
1.2.1 Physical Vapor Deposition

PVD methods (which include sputtering, electron-beam evaporation, cathodic-arc deposition, resistive-heating evaporation, and pulsed-laser deposition) involve the generation of a vapor flux by raising a bulk material to a sufficiently high energy level so that particles known as adatoms (clusters of atoms) are ejected from the bulk material [13,14]. The goal is for the adatoms to form a collimated vapor flux that reaches the substrate and condenses thereupon.

In order to control film morphology, adatoms need to arrive at the substrate surface in a predictable manner. This is achieved through increasing the mean free path of the adatoms to orders of magnitude more than the dimensions of the PVD chamber by reaching vacuum levels of $10^{-3}$ to $10^{-6}$ Torr for resistive-heating evaporation and $10^{-1}$ to $10^{-3}$ Torr for sputtering [2]. In addition, a high vacuum means that fewer impurities, such as atmospheric gases, water vapor, and dust accumulate on the surface or in the bulk of the growing film. During PVD, the adatoms leave the receptacle containing the source (bulk) material in a cosine-distributed fashion, as shown in Fig. 1.1. The combination of high vacuum and cosine distribution ensures that a large fraction of the adatoms will travel in a straight line from the source receptacle towards the substrate. This line-of-sight characteristic is advantageous over other thin-film methods such as chemical vapor deposition (CVD) [14]. In the case of sputtering, line-of-sight deposition is less easily achievable since some ionized gas needs to be introduced to knock off adatoms.
from the source material. However, using electric and magnetic fields, magnetron sputtering can allow for ionization at lower pressures, thereby increasing the mean free path of adatoms [2].

Since the vapor flux is not planar, there will be a slight variation in thickness radially from the center of the substrate. In order to reduce this effect, the substrate must be placed sufficiently far from the source receptacle in order to reduce the solid angle subtended by the substrate on the receptacle, which acts as a point source [2]. However, this must also be balanced with the fact that moving the substrate farther will reduce the maximum deposition rate that can be achieved. One benefit that oblique-angle deposited films have is that the solid angle decreases as the angle between the average direction of the vapor flux and the substrate normal increases, further planarizing the vapor flux.

**Figure 1.1.** Different cosine distributions can be achieved using different source receptacles and are also dependent of the specific type of PVD [2].
1.2.2 Salient Developments in CTF History

The optical properties of thin films were first noticed and studied by Faraday in 1857, who used thermal evaporation to make his films [15]. The first person to seemingly notice the non-uniformity of PVD when he deposited metal on a glass slide was Wright, who placed a metal loop about 3–4 mm in diameter a distance of 3 mm away from a 30-mm-diameter glass slide [16]. The proximity of the loop and its smaller size led to a large sweeping of the polar angle in order to cover the whole substrate surface. Due to the cosine-distributed emission of adatoms with respect to the azimuthal angle, it is no wonder that his films possessed a radial non-uniformity. As he desired to fabricate a metal mirror, uniformity was required and so he devised a way to move the substrate with respect to the loop in order to average the vapor flux direction over the whole surface.

Later, in 1886, Kundt, using a similar deposition setup, confirmed the radial non-uniformity. He went one step further and conjectured that the optical anisotropy was due to anisotropy in the way the material had been deposited [2,17]. At this time, it was hard to prove the validity of this statement, as the necessary technologies for scanning electron microscopy (SEM) would not be available until many decades later. Thanks to SEM, the crucial discovery of self-shadowing was made by König and Helwig [18], [2]. They realized that under certain conditions when the adatom mobility on the substrate surface was low, it was possible for some features of the growing film to block other parts of the growing films (or
the substrate during very early growth) from the incoming vapor flux. This self-shadowing phenomenon is possible due to the line-of-sight characteristic of PVD.

Once adatoms arrive on the surface of the substrate, the process of their sticking to the film is determined by the ratio of the substrate temperature $T$ to the melting temperature $T_m$ of the bulk material [2]. If the reduced temperature $T/T_m$ is low, the adatoms stay locally where they first arrive. At a very high reduced temperature, the adatoms may adsorb and then desorb from the substrate surface. In between these two extremes, the adatoms can adsorb to the substrate and then diffuse along the surface or into the bulk of the film. This ability of the adatoms to move once adsorbed is characterized by the adatom mobility. Depending on the parameters of the deposition, these behaviors prevent the growth of a smooth planar film and instead cause the adatoms to cluster so that the film grows with a conical or columnar morphology. This phenomenon was first documented by Movchan and Demchishin in 1962 [19] and its physical characterization later improved by Thornton [20] and Messier and Troiler-McKinstry [21].

1.2.2.1 Structure Zone Model

Movchan and Demchishin created what is now known as the Structure Zone Model (SZM), to pictorially describe the film morphology as a function of $T/T_m$ [2], [21]. After studying thin films grown by electron-beam evaporation, Movchan and Demchishin identified Zones 1, 2, and 3. At low reduced temperature ($T/T_m < 0.3$) [20],
the adatoms arrive in 1–3-nm clusters and essentially do not move after adsorption. Since there is a distribution of sizes, larger adsorbed clusters will have a better chance of capturing incoming adatoms. This process repeats and engenders competitive growth, whereby large clusters surpass small ones and continue to grow in a conical form. This type of growth occurs in Zone 1 of the SZM, as illustrated in Fig. 1.2. Zone 2 occurs at higher reduced temperatures \((0.3 < T/T_m < 0.5)\) [20] when the adatoms have significant surface diffusion. Here, straight-sided crystalline grains are formed. As the temperature increases further \((0.5 < T/T_m < 1)\) [20], bulk diffusion into the film dominates and the columns disappear, being replaced with equiaxed crystals. This morphology is characteristic of Zone 3.

After studying the morphology of thin films made by magnetron sputtering, Thornton added another parameter to the SZM. This parameter was the background gas pressure, and its incorporation led to the identification of Zone T ("T" for transition) between Zones 1 and 2. It is here that the growth of clusters starts to become non-competitive, and the morphology is of poorly defined fibrous grains without void boundaries [22].

Finally, Messier and Trolier-McKinstry realized that with low ion-bombardment at higher pressures, a part of Zone T gains new characteristics. Therefore they inserted Zone M between Zones 1 and T [21]. In Zone M, the grains are better defined, with no tapering as there was in Zone 1, but instead resemble matchsticks. As growth occurs, ion-bombardment helps sputter adatoms to what
would be smaller clusters that would otherwise be shadowed by the larger clusters. This allows for non-competitive growth of columns with a surrounding void space. This is the desired zone for growing CTFs, and is also the appropriate zone to grow STFs [2].

**Figure 1.2.** The five possible zones of PVD in the SZM. Zone M is the correct zone for growing CTFs and STFs [2].

### 1.2.2.2 CTF Growth

Prior to growing STFs, one must be able to grow CTFs. As stated in Sec. 1.2.2.1, it was found that non-competitive columns will grow in Zone M with low-energy-assisted ion-bombardment. The propensity toward column morphology is accentuated by directing the vapor flux obliquely towards the substrate; the angle between the vapor-flux direction and the substrate plane being denoted by $\chi_v \in (0^\circ, 90^\circ]$ in
Then, as clusters nucleate and grow, they block the area directly behind from arriving adatoms. This self-shadowing tends to work well for $\chi_v \ll 90^\circ$. The columns grow at an angle $\chi$ often related to $\chi_v$ by

$$\tan(\chi) = m \tan(\chi_v),$$  \hspace{1cm} (1.1)

where $m$ is a parameter specific to the material being deposited found from empirical data [23], and $\chi \geq \chi_v$ [2]. The angle $\chi$ can be measured using cross-sectional SEM images.

### 1.2.3 STF History and Growth

To change from columnar growth to more complicated but still locally columnar growth, all that was needed was to instantaneously change the direction of the vapor flux during deposition. In 1966, Nieuwenhuizen and Haanstra attempted to grow a cascade of two CTFs with a sudden change in $\chi_v$, the resulting mor-
phology comprising of chevronic nanocolumns [24]. This was a key step towards the eventual emergence of STFs. Motohiro and Taga made a Ta$_2$O$_5$ quarter-wave retardation film with nanocolumns of chevronic shape [25]. The difference from Nieuwenhuizen and Haanstra’s experiment is that Motohiro and Taga changed the vapor-flux direction several times.

However, the first to make chiral STFs were Young and Kowal [26] who deposited fluorite films while slowly and continuously rotating the substrate about a central normal axis. At the time, SEM imaging was not widely available; yet they speculated that remarkable optical properties must arise from, “helically symmetrical arrangement of crystallites, crystal growth or voids” [26]. From these experiments, it was imagined that a wide range of morphologies could be possible by changing the angle $\chi_v$ [2,27]. These morphologies can fall into two categories: nematic and helical [2]. Sculptured nematic thin films (SNTFs) consist of quasi-2D structures such as slanted columns, chevrons, zig-zags, and C- and S-shaped nanocolumns, as exemplified in Fig. 1.4. Thin-film helicoidal bianisotropic mediums (TFHBMs) consist of nanohelixes and super-nanohelixes and are commonly referred to as chiral STFs; see Fig. 1.5.

1.2.3.1 Evolution of STF Growth Methods

Several methods have been devised to grow complex STFs. The first attempt was by Robbie et al. [28] who used two evaporation sources to deposit chevronic
columns of SNTFs. In order to achieve vapor-flux angles $\chi_v \in [5^\circ, 15^\circ]$ the substrate had to be placed close to the receptacle, which imposed two problems. The close proximity to the source heated the substrate, thereby increasing adatom mobility as became evident by the lack of porosity. The close proximity also allowed for a greater vapor-flux gradient across the substrate, yielding a non-uniform film. Because of these two problems, a different method was attempted. This involved one source and rotating the substrate periodically by $180^\circ$ about its central normal axis. A large vapor source was used to ensure symmetric vapor fluxes at both
deposition positions. Although an improvement, these two methods were only capable of creating SNTFs, not chiral STFs.

Unknowingly, the method of Young and Kowal [26] was rediscovered by Robbie et al. [30] to deposit chiral STFs. Their idea was to deposit with one source and have the substrate rotate continuously and slowly about its central normal axis. With the ability to characterize the morphology using an SEM, they confirmed the formation of nanohelixes, thereby vindicating Kowal and Young [26]. Instead of continuous rotation, the substrate is often rotated in small angular increments $\delta$ about the central normal axis, as shown in the first row of Figure 1.6. The resulting chiral STF may be called a finely ambichiral thin film [31].

Hodgkinson et al. [32] proposed the serial bideposition (SBD) method to make chiral STFs [32, 33], where deposits are made $180^\circ + \delta$ apart and the process is repeated until a full revolution is complete, as shown in the second row of Figure 1.6. With this, Hodgkinson et al. were able to observe a significant enhancement of the CBP at $\theta_{inc} = 0^\circ$ exhibited by thin chiral STFs.

A further modification to the SBD method came from Pursel and Horn [11]. Two subdeposits would be made $180^\circ$ from each other before the small angle $\delta$ was introduced, as shown in the third row of Figure 1.6. With the modified SBD method [11], the subdeposit heights are symmetric. This naturally leads to the question of what would happen if the subdeposit heights were asymmetric, also first explored by Pursel and Horn [11]. They compared two chiral STFs grown at
Figure 1.6. Four schemes of substrate rotation to grow chiral STFs. In these schemes, the subdeposit structure is divided differently [11].

\( \chi_v = 12.5^\circ \) and \( \delta = 1^\circ \), the first with a 1:1 subdeposit-height ratio and the second with a 3:1 subdeposit-height ratio. They found that their ASBD method still led to the exhibition of the CBP by the chiral STF while possessing different void-network characteristics. In regards to the optics of ASBD films, three main results were found: (1) the non-co-handed transmittance of the 3:1 film was slightly less than that of the 1:1 film, (2) the non-co-handed transmittances at lower wavelengths of the 3:1 film was less than that of the 1:1 film, and (3) the central wavelength red-shifted from \( \lambda_o^{Br} = 539 \) nm for the 1:1 film to \( \lambda_o^{Br} = 594 \) nm for the 3:1 film.
1.3 Motivation

Pursel and Horn proved that ASBD could preserve the CBP. The work done for this thesis systematically explores the dependence of the CBP on the asymmetry in the ASBD technique, with hopes of improving the exhibition of the CBP by chiral STFs. Hence, chiral STFs with subdeposit-height ratios 1:1, 1.5:1, 2:1, 2.5:1, 3:1, 5:1, 7:1, and 9:1 were deposited along with a finely ambichiral sample. Furthermore, this work expands upon Pursel and Horn’s research by characterizing how the asymmetry will change the CBP for $\theta_{inc} > 0^\circ$. Ideally, one would like to increase the effectiveness of the CBP for circular-polarization filters for a large range of $\theta_{inc}$. 
Chapter 2  |  

The Circular Bragg Phenomenon  

2.1 The Bragg Phenomenon  

Preceding the CBP was the Bragg phenomenon (BP), which was proposed by William Henry Bragg and William Lawrence Bragg in 1915 [34]. The BP occurs when electromagnetic waves within a certain spectral regime centered around a central wavelength \( \lambda_{Bo} \) get almost totally reflected from the surface of a crystal. The same BP is exhibited by a multi-layered material with layers of alternating high/low indices of refraction. Each high/low-index layer needs to be one-quarter of the wavelength desired to be highly reflected. Different bands of reflection exist
with their center wavelengths given by [1].

\[ \lambda_{om}^{Br} = \frac{4\Omega}{m} n_{avg} \cos \theta_{inc}, \quad m \in \{1, 2, 3, \ldots\}, \quad (2.1) \]

where \( 2\Omega \) is the period of the index of refraction, \( n_{avg} \) is the spatially averaged index of refraction, and \( \theta_{inc} \) is the angle of incidence of the electromagnetic plane wave with respect to the thickness direction. It is assumed for Eqn. (2.1) that the materials are non-dispersive. For multi-layers made of isotropic materials, the BP is insensitive to the polarization state of the incident electromagnetic wave in the neighborhood of \( \lambda_{om}^{Br} \) [1].

Discrimination between the two linearly polarized states can be forced by simply making one or both layers out of a uniaxial dielectric material [2]. The peak reflectances for incident \( p \)- and \( s \)-polarized light are then centered around different wavelengths. To discriminate even further, the ordinary and extraordinary refractive indices, \( n_o \) and \( n_e \), of the uniaxial material can be chosen such that incident light of one state of polarization is almost totally transmitted while that of the other is highly reflected [13]. This occurs under the conditions \( \{n_{oa} \neq n_{ob}, \ n_{ea} = n_{eb}\} \) or \( \{n_{oa} = n_{ob}, \ n_{ea} \neq n_{eb}\} \), where \( n_{oa}, n_{ob}, n_{ea}, \) and \( n_{eb} \) are the ordinary refractive index of layer a, ordinary refractive index of layer b, extraordinary refractive index of layer a, and extraordinary refractive index of layer b, respectively. One or both layers could be made of a biaxial dielectric material. A periodic multi-layer is also called a Bragg mirror or a Bragg reflector.
2.2 Reusch Piles

In a Reusch pile, a period consists of multiple layers of the same uniaxial dielectric material such that the optic axis of every uniaxial layer is rotated in relation to that of the previous uniaxial layer by a constant angle about the thickness axis. It was found that a Reusch pile selectively discriminates between circularly polarized states of normally incident light in multiple spectral regimes [1,35–39]. For normally incident light and for the optic axis in each layer being normal to the thickness direction, and with the assumption of the material being non-dispersive, the circular Bragg regimes will be centered around

$$\lambda_{Br}^{pq} = \frac{2\Omega(n_o + n_e)}{pq + 1}, \quad p \in \{0, 1, 2, 3, \ldots\},$$  \hspace{1cm} (2.2)

where \( q \) is the number of layers in a half-period, \( 2\Omega \) is the period, and \( n_o \) and \( n_e \) are the ordinary and extraordinary refractive indices, respectively. In addition, the uniaxial material can be replaced by a biaxial material. CTFs can be incorporated in Reusch piles, where they have been found experimentally to exhibit two identifiable circular Bragg regimes [31,37]. As \( q \to \infty \), the changing orientation of the permittivity dyadic from layer to layer is essentially continuous on a macroscopic scale and the CTF can be considered to be a chiral STF [31].
2.3 Cholesteric Liquid Crystals

The permittivity dyadic of a chiral liquid crystal (CLC) varies continuously about a fixed axis. CLCs possess short-range order like a liquid material, yet still possess long-range periodicity of solid crystals [40]. CLCs fall into two categories: nematic and smectic. Both chiral nematic liquid crystals (also called cholesteric liquid crystals) and chiral smectic liquid crystals exhibit the CBP. These materials can also be thought of as Reusch piles in the limit \( q \to \infty \) [1].

2.4 Chiral STFs

Simililar to CLCs, chiral STFs can be thought of as Reusch piles in the limit of \( q \to \infty \). The relative permittivity dyadic of a chiral STF is given by [2]

\[
\varepsilon_r = S \cdot \varepsilon_{ref} \cdot S^{-1}, \tag{2.3}
\]

where

\[
\varepsilon_{ref} = \begin{pmatrix} 
\varepsilon_b & 0 & 0 \\
0 & \varepsilon_c & 0 \\
0 & 0 & \varepsilon_a 
\end{pmatrix}, \tag{2.4}
\]
with $\varepsilon_a$, $\varepsilon_b$, and $\varepsilon_c$ being the principal relative permittivity scalars of the biaxial dielectric material. The helical morphology is captured by the rotation dyadic

$$S = \begin{pmatrix}
\cos(hz \frac{\pi}{\Omega}) & -\sin(hz \frac{\pi}{\Omega}) & 0 \\
\sin(hz \frac{\pi}{\Omega}) & \cos(hz \frac{\pi}{\Omega}) & 0 \\
0 & 0 & 1
\end{pmatrix}
\begin{pmatrix}
\cos(\chi) & 0 & -\sin(\chi) \\
0 & 1 & 0 \\
\sin(\chi) & 0 & \cos(\chi)
\end{pmatrix}.$$  \( (2.5) \)

Here, $h$ can take on the value 1 or $-1$ for structural right or left handedness respectively, and $\chi$ is the angle of nanocolumn growth with respect to the substrate. Not only do $\varepsilon_a$, $\varepsilon_b$, and $\varepsilon_c$ depend on the material evaporated to fabricate the chiral STF, but also on $\chi$ and $2\Omega$.

The CBP for a chiral STF occurs in spectral regimes with center wavelengths \[41\], \[42\]

$$\lambda_{bm}^{Br} \approx \frac{2\Omega}{m} (\sqrt{\varepsilon_c} + \sqrt{\varepsilon_d}) \cos^{1/2} \theta_{inc}, \ m \in \{1, 2, 3, ...\}.$$  \( (2.6) \)

where \[43\]

$$\varepsilon_d = \frac{\varepsilon_a \varepsilon_b}{\varepsilon_a \cos^2 \chi + \varepsilon_b \sin^2 \chi}$$  \( (2.7) \)

and the absence of dispersion is assumed. Only orders $m > 1$ will clearly distinguish between incident RCP and LCP plane waves. The order $m = 2$ is the most important one, for which the bandwidth is

$$\Delta\lambda_0^{Br} \approx 2\Omega \left| \sqrt{\varepsilon_c} - \sqrt{\varepsilon_d} \right| \cos^{1/2} \theta_{inc}.$$  \( (2.8) \)
and the center wavelength

\[ \lambda_o^{Br} \approx \Omega (\sqrt{\varepsilon_c} + \sqrt{\varepsilon_d}) \cos^{1/2} \theta_{inc} \]  

(2.9)

It is important to note that this model assumes the chiral STF to be a material continuum; therefore it does not explicitly consider scattering of light due to the void network.
Chapter 3

Experimental Procedure

3.1 PVD Method

The deposition method used to grow all films for this thesis was resistive-heating thermal evaporation, which is typically used for materials with vaporizing temperatures below 1500 °C [44]. In this method, a current is passed through a tungsten boat containing the bulk (source) material to be evaporated. In addition to a thermal-evaporation system being readily available at Pennsylvania State University, advantages of thermal evaporation include the ability to evaporate bulk material at high vacuum, high deposition rates, relatively low power input, and directionality. Figure 3.1 is a photograph of the vacuum chamber used.
3.2 Material Choice

99.995% pure zinc selenide (ZnSe) supplied by Alfa Aesar® (Ward Hill, MA, USA) was chosen for evaporation. The lumps of ZnSe were crushed into a fine powder using a mortar and pestle. A respirator was worn during the experiments to avoid the toxic effects of ZnSe on the respiratory system [45]. The melting point of
ZnSe has been reported to be 1525 °C at 1 atm pressure, and will be lower under vacuum conditions [45,46]. Kurt J. Lesker, Inc. (Jefferson Hills, PA, USA) reports the vaporization pressure of ZnSe at $10^{-4}$ Torr to be 660 °C [47]. This value will further decrease due to the order-of-magnitude lower-vacuum conditions during deposition, making the evaporation of ZnSe well within the capabilities of resistive-heating thermal evaporation. ZnSe was also chosen for its high index of refraction ($n = 2.67$ at $\lambda_o = 550$ nm) which is important for increasing the local linear birefringence of the chiral STF [32]. Various companies report high transmission for ZnSe over a wide range of wavelengths that cover the visible spectrum [48,49]. ZnSe is a II-VI compound semiconductor with a bandgap of 2.71 eV [50].

### 3.3 Substrate Preparation

Each chiral STF was deposited simultaneously on a glass substrate and silicon substrate. VWR® pre-cleaned micro-slides (48300-0025, VWR, Radnor, PA, USA) were cut into smaller pieces. The glass substrate was used for the sample to be optically characterized and the silicon substrate for SEM imaging of the sample. Both substrates were first cleaned by hand with ethanol and then bathed in ethanol while in an ultrasonic cleaner for 5 min on each side. Immediately after removal from the bath, the substrates were dried with pressurized nitrogen gas, secured to the substrate holder, and placed in the vacuum chamber. Kapton® tape was used to secure the substrates to the substrate holder.
3.4 Deposition Procedure

All chiral STFs were fabricated approximately 10 periods thick, each period being approximately 300 nm. The vapor-flux incidence angle was set as $\chi_v = 20^\circ$ along with $\delta = 3^\circ$. Due to the size limitations of the tungsten boat (S22-.005W, R. D. Mathis, Signal Hill, CA, USA), the process was completed in three stages, each depositing 1000 nm. In between two successive stages, the chamber was cooled and evacuated to allow the replacement of source material. In all cases, the chamber was allowed to cool for half-an-hour before venting to prevent thermal stresses on the film. Care was taken not to alter the orientation of the substrate holder in between two consecutive stages.

To monitor the deposition rate, a gold-coated quartz crystal monitor (QCM) was used. Once placed in the probe, an oscillator circuit runs at the resonance frequency of the crystal (6 MHz) and induces shear oscillations of the QCM volume which in turn causes a detectable piezoelectric effect that is used to monitor the rate. Every crystal (SPC-1093-G10, Inficon, Chicago, IL, USA) was discarded once it reached 80% of its lifetime. To ensure the QCM was accurate, the tooling factor was adjusted and the correct $z$-ratio was selected.

Since the QCM cannot be at the same position as the substrate, the QCM and substrate will receive different amounts of the vapor flux. In order to account for this difference, the thickness of a film must be measured by some other means,
such as a stylus or optical profilometer. The thickness read by the QCM is then corrected to the measured thickness using the relation

$$T_f = T_i \frac{t_m}{t_{QCM}},$$

(3.1)

where $T_f$, $T_i$, $t_m$, and $t_{QCM}$ are the final tooling factor, initial tooling factor, measured thickness, and thickness read by the QCM, respectively.

While the tooling factor accounts for the correct amount of material on the QCM, the $z$-ratio is a measure of how much the depositing material will impede the QCM resonance oscillations. Here, $z$ is the specific acoustic impedance (kg/m$^2$s) of a material, and the $z$-ratio is the ratio of the specific acoustic impedance of the quartz crystal to the specific acoustic impedance of the source material. ZnSe has a $z$-ratio of 0.722 [51].

Once both substrates were taped to the substrate holder which was mounted onto the motor, the door of the vacuum chamber was closed and the chamber was pumped down. Throughout each deposition, the pressure was kept below $10^{-5}$ Torr. Leading up to the deposition, the current was slowly turned up to heat the tungsten boat. The current was increased at approximately 0.1 A/s until the voltage was $\sim 2$ V and the electrical power supplied to the boat was $\sim 190$ W. This slow ramping rate was a safeguard to make sure that material would not be ejected from the boat from too sudden a power increase. This also allowed the material to outgas so that the actual deposition could take place under
higher vacuum. In addition, this step allowed the material to sinter homogeneously, thereby preventing large portions of the material to sinter during deposition and causing spikes in the deposition rate. Lastly, any contaminants on the boat or in the source material would have had sufficient time to vaporize before the deposition. The deposition took place at 0.4 ± 0.02 nm/s. Once this rate was reached, the shutter was manually opened, the motor program was manually started, and the QCM thickness reading was zeroed.

3.5 Motor Program

Two motors, the first to orient the substrate holder to the chosen value of $\chi_v$ and the second to rotate the substrate about a central normal axis, were controlled by an Applied Motion© SiNet Hub 444 Programmer (1000-267, Applied Motion Products, Watsonville, CA, USA). Before the chamber was pumped, the first motor was run for visual confirmation that the substrate was oriented correctly towards the source. The second motor was programmed to automatically stop after $10/3$ periods. The rotating motor must stop for each subdeposit to grow. The wait times for consecutive subdeposits were determined by a Mathematica™ program; a sample is shown in Fig. 3.2.

A trial run was done to ensure that the second motor stopped after the correct amount of time and in the correct position for the next subdeposit. It was found that the motor stopped in the correct place but would have rotated for an addi-
tional ∼ 1 min while 10/3 periods had been deposited. This extra time could be attributed to two reasons. First, the SiNet Hub Programmer has one caveat, which is that only certain values for time intervals can be inputed. Since any inputed time interval never exactly matched the calculated time interval, any slight error would be multiplied over the course of 400 subdeposits. The second error could come from delay time in between motor steps. The error of ∼ 1 min for each deposition at a rate of 0.4 nm/s would amount to a total thickness of 3075 nm. The discrepancy of 3075 − 3000 = 75 nm is within the 5% error limit that has been deemed acceptable from previous work [4], [52], [53]. Due to manual control of the deposition rate during the deposition, the targeted 0.4 nm/s was not always achieved, and therefore the thickness from sample to sample varied slightly from 3075 nm. Even with a slight error in pitch, all that matters is that each sample was fabricated in the same fashion.
Figure 3.2. The MMI Prompt commands were used in the Applied Motion SiNet Hub Programmer to pause at critical points until manual input was given to continue. Also in the bottom left corner is the analysis window, from which the wait times were inputed into a Mathematica™ provided in Fig. 3.3.
Figure 3.3. Mathematica\textsuperscript{TM} program for the 3:1 film. The values of the wait-time ratio $n$ were changed for different $h_1/h_2$ ratios. The outputs show what values to enter for the subdeposit wait times from Figure 3.2 and how many program repeats the motor needs.
3.6 Characterization

3.6.1 Optical Characterization

All samples were optically characterized within 24 h of fabrication. This was to ensure that water adsorption and consequent degradation would not have an effect on transmittances and reflectances. The samples were kept in a desiccator from when they were fabricated up until the moment of characterization.

Light from a halogen source (HL-2000, Ocean Optics, Dunedin, Florida, USA) was passed through a fiber optic cable and then through a linear polarizer (GT10, ThorLabs, Newton, NJ, USA), a Fresnel Rhomb (LMR1, ThorLabs), the sample to be characterized, a second Fresnel Rhomb (LMR1, ThorLabs), a second linear polarizer (GT10, ThorLabs), and then a fiber optic cable to a CCD spectrometer (HRS-BD1-025, Mightex Systems, Pleasanton, CA, USA). Measurements were taken for the transmittance $T_{LL}$, $T_{RL}$, $T_{RR}$, and $T_{LR}$ for $\theta_{\text{inc}} \in [0^\circ - 70^\circ]$, and for the reflectance $R_{LL}$, $R_{RL}$, $R_{RR}$, and $R_{LR}$ for $\theta_{\text{inc}} \in [10^\circ - 70^\circ]$. The first subscript denotes the circular polarization state of light collected by the detector, while the second subscript denotes the circular polarization state of light impinging on the sample.
Figure 3.4. Photograph of the setup to measure the circular transmittances of a sample.

Figure 3.5. Photograph of the setup to measure the circular reflectances of a sample.
In order for the sample to rotate about the center, stage 1 in Figs. 3.4 and 3.5 was rotated 90° and the sample was moved until half of the initial intensity was observed by the detector. Then, stage 1 was rotated back so that the normal to the substrate was more or less in the direction of the incoming beam. With an index card, the reflected light from the sample was checked against the incoming beam. Stage 1 was then further adjusted so that these two beams overlapped. The position of stage 1 was then zeroed. These steps ensured that the substrate rotation and the stage rotation with respect to the incoming light would be the same. All data were taken in a dark room to avoid noise from external sources. The data were normalized as follows:

\[
T_{LL_{\text{relative}}} = \frac{T_{LL_{\text{measured}}} - I_{dark}}{I_{LL} - I_{dark}}
\]

(3.2)

\[
T_{LR_{\text{relative}}} = \frac{T_{LR_{\text{measured}}} - I_{dark}}{I_{LL} - I_{dark}}
\]

(3.3)

\[
T_{RL_{\text{relative}}} = \frac{T_{RL_{\text{measured}}} - I_{dark}}{I_{RR} - I_{dark}}
\]

(3.4)

\[
T_{RR_{\text{relative}}} = \frac{T_{RR_{\text{measured}}} - I_{dark}}{I_{RR} - I_{dark}}
\]

(3.5)

where \(T\) can be replaced with \(R\) for the reflectance data. \(I_{LL}\) and \(I_{RR}\) are the intensities of LCP and RCP, respectively, without the sample present and \(I_{dark}\) is the intensity in the absence of the white light source and background light sources.
3.6.2 Cross-section Morphology

The cross-section morphology was characterized using a FEI Nova™ NanoSEM 630 (FEI, Hillsboro, Oregon, USA). FESEM works by applying an electric field to the tip of a sharpened tungsten filament, which allows for the emission of electrons from a smaller area with a more narrow range of energies. This increases the resolution compared to a thermionic emission SEM. FESEM images allowed for the comparison of the chiral morphology and void network from sample to sample. The freeze-fracture technique was used on films deposited on Si for edge viewing. This technique has shown to be delicate enough to preserve biological samples [54]. All samples were sputtered with iridium using a Quorum Emitech® K575X (Quorum Technologies Ltd., Ashford, Kent, United Kingdom) sputter coater.
Chapter 4  

Results

As mentioned in Chapter 3, nine samples were made with subdeposit heights in the ratios described in Table 4.1. For all samples except the finely ambichiral sample, the total height $h_{x:1} = h_1 + h_2$ of two consecutive subdeposits was kept constant. For the ambichiral sample, the deposition procedure followed that of the

<table>
<thead>
<tr>
<th>Ratio</th>
<th>$h_1$ (nm)</th>
<th>$h_2$ (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1:1</td>
<td>1.08</td>
<td>1.08</td>
</tr>
<tr>
<td>1.5:1</td>
<td>1.30</td>
<td>0.86</td>
</tr>
<tr>
<td>2:1</td>
<td>1.44</td>
<td>0.72</td>
</tr>
<tr>
<td>2.5:1</td>
<td>1.55</td>
<td>0.62</td>
</tr>
<tr>
<td>3:1</td>
<td>1.62</td>
<td>0.54</td>
</tr>
<tr>
<td>5:1</td>
<td>1.81</td>
<td>0.36</td>
</tr>
<tr>
<td>7:1</td>
<td>1.90</td>
<td>0.27</td>
</tr>
<tr>
<td>9:1</td>
<td>1.95</td>
<td>0.21</td>
</tr>
<tr>
<td>Finely ambichiral</td>
<td>2.49</td>
<td>N/A</td>
</tr>
</tbody>
</table>

Table 4.1. First and second subdeposit heights and their ratio. The deposition procedure followed for samples 1:1 to 9:1 is shown in the fourth row of Fig. 1.6. The finely ambichiral sample followed the deposition procedure in the first row of Fig. 1.6.

The first row in Figure 1.6. Since the rotation sequence of the two procedures differed,
the subdeposit height of the ambichiral sample had to be altered to $h = 2.49$ nm in order to grow the same number of periods within the same thickness.

### 4.1 Optical Responses

As stated in Chapter 3, all films were illuminated by LCP and RCP plane waves and the circular remittances were measured. The raw data were exported in an Excel$^{TM}$ worksheet. Due to the large size of each Excel$^{TM}$ file, the data were compiled using a Mathematica$^{TM}$ program, which is provided in Appendix B. A sample of the raw data is provided in Appendix A. Due to the limitations of the optical setup, circular reflectances were measured for $\theta_{\text{inc}} \geq 10^\circ$. Good predictions for normal reflectance can be made from data for $\theta_{\text{inc}} = 10^\circ$ [4].

#### 4.1.1 Density Plots of Circular Remittances

All circular remittance were plotted as functions of $\lambda_o$ and $\theta_{\text{inc}}$ in 2D-density plots in Figures 4.1–4.18. For a desirable circular-polarization filter, $R_{RR} - R_{LL}$ and $T_{LL} - T_{RR}$ should all be close to unity, while $R_{RL}, R_{LR}, T_{RL},$ and $T_{LR}$ should be vanishingly small. By virtue of the principle of conservation of energy, no circular remittance can exceed unity.
Figure 4.1. Measured circular reflectances of the 1:1 sample.

Figure 4.2. Measured circular transmittances of the 1:1 sample.
Figure 4.3. Measured circular reflectances of the 1.5:1 sample.

Figure 4.4. Measured circular transmittances of the 1.5:1 sample.
Figure 4.5. Measured circular reflectances of the 2:1 sample.

Figure 4.6. Measured circular transmittances of the 2:1 sample.
Figure 4.7. Measured circular reflectances of the 2.5:1 sample.

Figure 4.8. Measured circular transmittances of the 2.5:1 sample.
Figure 4.9. Measured circular reflectances of the 3:1 sample.

Figure 4.10. Measured circular transmittances of the 3:1 sample.
Figure 4.11. Measured circular reflectances of the 5:1 sample.

Figure 4.12. Measured circular transmittances of the 5:1 sample.
Figure 4.13. Measured circular reflectances of the 7:1 sample.

Figure 4.14. Measured circular transmittances of the 7:1 sample.
Figure 4.15. Measured circular reflectances of the 9:1 sample.

Figure 4.16. Measured circular transmittances of the 9:1 sample.
Figure 4.17. Measured circular reflectances of the finely ambichiral sample.

Figure 4.18. Measured circular transmittances of the finely ambichiral sample.
Two main qualitative conclusions can be deduced from Figs. 4.1–4.16. First, $\lambda_o^{Br}$ redshifts for all $\theta_{inc}$ as the ratio $h_1/h_2$ increases. Second, the circular Bragg regime narrows for all $\theta_{inc}$ with increasing ratio $h_1/h_2$.

Starting with the 1:1 sample, Figs. 4.1 and 4.2 show that the circular Bragg regime is clearly identifiable for low values of $\theta_{inc}$, but does not manifest clearly for $\theta_{inc} \gtrsim 60^\circ$. This can be seen from the magnitude of the difference $R_{RR} - R_{LL}$ in the circular Bragg regime. The plots of $T_{LR}$ and $T_{RL}$ in Fig. 4.2 show that some cross-polarization occurs on transmission in the circular Bragg regime, an unwanted effect for circular-polarization filters.

Figures 4.3 and 4.4 for the 1.5:1 samples show considerable increase in $R_{RR} - R_{LL}$ at $\theta_{inc}$ above $60^\circ$. However, discrimination between incident LCP and RCP plane waves is still not as strong as it is at lower $\theta_{inc}$. The difference $T_{LL} - T_{RR}$ intensifies at higher angles of incidence for the most part of the spectrum: about 20% higher than what $T_{LL} - T_{RR}$ is for the 1:1 sample.

The remittances of sample 2:1 are curious in relation to all other asymmetric samples. This sample possesses a well-defined circular Bragg regime with an improvement in $R_{RR} - R_{LL}$ at higher $\theta_{inc}$ compared to the 1:1 and 1.5:1 samples. However, $T_{LL} - T_{RR}$ within the circular Bragg regime is lower compared to the 1:1 and 1.5:1 samples. From the pattern formed by the other asymmetric samples, there does not seem to be any reason why the 2:1 sample should inherently possess decreased transmittance discrimination. This may be due to an obstruction on the
surface of the film during optical characterization. It was determined that it was unnecessary to fabricate this film again, as enough data on how ASBD affects the CBP was extracted from the other eight samples.

Red-shifting and narrowing of the circular Bragg regime becomes very clear when comparing the 3:1 sample (Figs. 4.9 and 4.10) to the 1:1 sample (Figs. 4.1 and 4.2). The 3:1 sample continues the trend of decreased cross-polarized transmittances in the circular Bragg regime.

From Figs. 4.11, 4.13, and 4.15, it becomes evident that the difference $R_{RR} - R_{LL}$ in the circular Bragg regime decreases as the ratio $h_1/h_2$ gets significantly high, especially for lower $\theta_{inc}$ for the 7:1 and 9:1 samples. A slight decrease in cross-polarization transmittance, however, continues to occur.

Since Figs. 4.13–4.16 did not differ drastically in terms of the red-shifting of the circular Bragg regime, a limit could have been reached. To confirm this conjecture, a finely ambichiral film was grown. The conjecture that the limit of the red-shifting of the circular Bragg regime was reached with the 9:1 sample turned out to be correct, since the profiles of the circular Bragg regime for the finely ambichiral sample (Fig. 4.18) and the 9:1 sample (Fig. 4.16) overlap almost perfectly. The difference $R_{RR} - R_{LL}$ for the finely ambichiral sample is significantly weaker in the circular Bragg regime compared to samples deposited with the ASBD method. Figures 4.11, 4.13, 4.15, and 4.17 suggest that if the ratio $h_1/h_2$ keeps increasing beyond 9:1, the circular Bragg regime will only discriminate strongly between $R_{RR}$
and $R_{LL}$ at increasingly higher $\theta_{inc}$. This means that an optimum value of the ratio $h_1:h_2$ exists. In addition, $\lambda_0^{Br}$ for all samples blue-shifts as $\theta_{inc}$ increases, which is in agreement with previous work [3, 4].

Figure 4.19. Blue-shifting of $\lambda_0$ for the 5:1 sample at various $\theta_{inc}$. All samples exhibited a similar blue-shifting effect.
4.1.2 Flatness Measurements

Instead of relying on color in the 2D-density plots to judge the quality of the CBP for each sample, a quantitative approach was taken. The figure of merit

\[ f(\lambda_o, \theta_{inc}) = 2 \frac{R_{RR}(\lambda_o, \theta_{inc}) - R_{LL}(\lambda_o, \theta_{inc})}{R_{RR}(\lambda_o, \theta_{inc}) + R_{LL}(\lambda_o, \theta_{inc})} \]  

(4.1)

has a value close to 2 if \( R_{RR} \) highly dominates \( R_{LL} \), which means that \( f(\lambda_o, \theta_{inc}) \) can be used as a measure of discrimination by the samples between incident LCP and RCP light.

Once the boundaries of the circular Bragg regime were found for \( \theta_{inc} \in [10^\circ, 70^\circ] \) at 10\(^\circ\) intervals, the quality of the CBP was determined by two parameters. The first parameter is the flatness of the CBP, and the second parameter is the magnitude of the difference between the average of \( f(\lambda_o, \theta_{inc}) \) within the circular Bragg regime and the ideal maximum value of 2.

The flatness of the CBP can be used to quantify how consistently the sample is discriminating throughout the entire circular Bragg regime for a fixed \( \theta_{inc} \). I defined the flatness function

\[ H(\theta_{inc}) = \frac{1}{\lambda_o^{max} - \lambda_o^{min}} \sum_{n=1}^{N} \left| f(\lambda_{o_n}, \theta_{inc}) - f_{avg}(\theta_{inc}) \right|^2 (\lambda_{o_{n+1}} - \lambda_{o_n}) , \]

(4.2)

where \( \lambda_o^{max} \) and \( \lambda_o^{min} \) are the upper and lower bounds of the circular Bragg regime.
respectively, $N$ is the number of wavelengths $\lambda_{o_n}$ selected for the averaging procedure, and

$$f_{avg}(\theta_{inc}) = \frac{1}{N} \sum_{n=1}^{N} f(\lambda_{o_n}, \theta_{inc}).$$

(4.3)

Figure 4.20 shows an example of a transmittance plot used to find the lower and upper bounds (points 1 and 2, respectively) of the circular Bragg regime for the 1:1 sample at $\theta_{inc} = 10^\circ$. Points 1 and 2 were also found from the reflectance plot. Points 1 and 2 from both plots were then averaged to find $\lambda_{o_{min}}$ and $\lambda_{o_{max}}$, respectively. The plots were made using the Mathematica$^TM$ code provided in Appendix D.
Figure 4.20. Plot of $T_{RR}$ of the 1:1 sample at $\theta_{inc} = 10^\circ$. The wavelength and transmittance at point 3 were found by averaging the respective values at points 1 and 2. Point 5 lies at the same wavelength as point 3, and the reflectance value was found by averaging the reflectance values of points 3 and 4.

The second parameter is defined as

$$g_{avg}(\theta_{inc}) = |f_{avg}(\theta_{inc}) - 2|. \tag{4.4}$$

If the CBP for a sample is ideally uniform (flat) and $R_{RR} - R_{LL}$ is high throughout the entire circular Bragg regime for a given $\theta_{inc}$, then $g_{avg}(\theta_{inc})$ will be close to zero. Tables 4.2–4.10 compile the data gathered from Eqns. (4.2) and (4.4). Figures 4.21 and 4.22 present plots of the tabled data points.
<table>
<thead>
<tr>
<th>$\theta_{inc}$</th>
<th>$H(\theta_{inc})$</th>
<th>$g_{avg}(\theta_{inc})$</th>
</tr>
</thead>
<tbody>
<tr>
<td>10</td>
<td>0.00131649</td>
<td>0.0277796</td>
</tr>
<tr>
<td>20</td>
<td>0.0171904</td>
<td>0.0566794</td>
</tr>
<tr>
<td>30</td>
<td>0.0389785</td>
<td>0.0988984</td>
</tr>
<tr>
<td>40</td>
<td>0.192069</td>
<td>0.256727</td>
</tr>
<tr>
<td>50</td>
<td>0.205673</td>
<td>0.450506</td>
</tr>
<tr>
<td>60</td>
<td>0.317899</td>
<td>0.943238</td>
</tr>
</tbody>
</table>

**Table 4.2.** Parameters to evaluate the quality of the CBP of the 1:1 sample as functions of $\theta_{inc}$.

<table>
<thead>
<tr>
<th>$\theta_{inc}$</th>
<th>$H(\theta_{inc})$</th>
<th>$g_{avg}(\theta_{inc})$</th>
</tr>
</thead>
<tbody>
<tr>
<td>10</td>
<td>0.0040231</td>
<td>0.0311214</td>
</tr>
<tr>
<td>20</td>
<td>0.0116021</td>
<td>0.0500576</td>
</tr>
<tr>
<td>30</td>
<td>0.0122508</td>
<td>0.0724109</td>
</tr>
<tr>
<td>40</td>
<td>0.0805326</td>
<td>0.198204</td>
</tr>
<tr>
<td>50</td>
<td>0.340013</td>
<td>0.534549</td>
</tr>
<tr>
<td>60</td>
<td>0.380371</td>
<td>0.9001</td>
</tr>
</tbody>
</table>

**Table 4.3.** Parameters to evaluate the quality of the CBP of the 1.5:1 sample as functions of $\theta_{inc}$.

<table>
<thead>
<tr>
<th>$\theta_{inc}$</th>
<th>$H(\theta_{inc})$</th>
<th>$g_{avg}(\theta_{inc})$</th>
</tr>
</thead>
<tbody>
<tr>
<td>10</td>
<td>0.00134901</td>
<td>0.0325292</td>
</tr>
<tr>
<td>20</td>
<td>0.000586236</td>
<td>0.0204816</td>
</tr>
<tr>
<td>30</td>
<td>0.00299498</td>
<td>0.0513688</td>
</tr>
<tr>
<td>40</td>
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<td>0.183174</td>
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<tr>
<td>50</td>
<td>0.261299</td>
<td>0.446261</td>
</tr>
<tr>
<td>60</td>
<td>0.338848</td>
<td>0.749292</td>
</tr>
</tbody>
</table>

**Table 4.4.** Parameters to evaluate the quality of the CBP of the 2:1 sample as functions of $\theta_{inc}$.
### Table 4.5. Parameters to evaluate the quality of the CBP of the 2.5:1 sample as functions of $\theta_{inc}$.

<table>
<thead>
<tr>
<th>$\theta_{inc}$</th>
<th>$H(\theta_{inc})$</th>
<th>$g_{avg}(\theta_{inc})$</th>
</tr>
</thead>
<tbody>
<tr>
<td>10</td>
<td>0.00396625</td>
<td>0.0357953</td>
</tr>
<tr>
<td>20</td>
<td>0.00331096</td>
<td>0.0340612</td>
</tr>
<tr>
<td>30</td>
<td>0.0187745</td>
<td>0.0812395</td>
</tr>
<tr>
<td>40</td>
<td>0.103959</td>
<td>0.210619</td>
</tr>
<tr>
<td>50</td>
<td>0.266196</td>
<td>0.451655</td>
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<tr>
<td>60</td>
<td>0.260344</td>
<td>0.712146</td>
</tr>
</tbody>
</table>

### Table 4.6. Parameters to evaluate the quality of the CBP of the 3:1 sample as functions of $\theta_{inc}$.

<table>
<thead>
<tr>
<th>$\theta_{inc}$</th>
<th>$H(\theta_{inc})$</th>
<th>$g_{avg}(\theta_{inc})$</th>
</tr>
</thead>
<tbody>
<tr>
<td>10</td>
<td>0.00216544</td>
<td>0.0225535</td>
</tr>
<tr>
<td>20</td>
<td>0.00367747</td>
<td>0.0292748</td>
</tr>
<tr>
<td>30</td>
<td>0.00942204</td>
<td>0.0699039</td>
</tr>
<tr>
<td>40</td>
<td>0.0557933</td>
<td>0.195999</td>
</tr>
<tr>
<td>50</td>
<td>0.0778055</td>
<td>0.345558</td>
</tr>
<tr>
<td>60</td>
<td>0.131491</td>
<td>0.628713</td>
</tr>
</tbody>
</table>

### Table 4.7. Parameters to evaluate the quality of the CBP of the 5:1 sample as functions of $\theta_{inc}$.

<table>
<thead>
<tr>
<th>$\theta_{inc}$</th>
<th>$H(\theta_{inc})$</th>
<th>$g_{avg}(\theta_{inc})$</th>
</tr>
</thead>
<tbody>
<tr>
<td>10</td>
<td>0.00492689</td>
<td>0.0227983</td>
</tr>
<tr>
<td>20</td>
<td>0.0012139</td>
<td>0.0212165</td>
</tr>
<tr>
<td>30</td>
<td>0.0053716</td>
<td>0.0669493</td>
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<tr>
<td>40</td>
<td>0.0469903</td>
<td>0.191825</td>
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<tr>
<td>50</td>
<td>0.09143</td>
<td>0.38757</td>
</tr>
<tr>
<td>60</td>
<td>0.316569</td>
<td>0.765968</td>
</tr>
<tr>
<td>$\theta_{inc}$</td>
<td>$H(\theta_{inc})$</td>
<td>$g_{avg}(\theta_{inc})$</td>
</tr>
<tr>
<td>----------</td>
<td>----------------</td>
<td>----------------</td>
</tr>
<tr>
<td>10</td>
<td>0.000492213</td>
<td>0.0244416</td>
</tr>
<tr>
<td>20</td>
<td>0.000703138</td>
<td>0.0325387</td>
</tr>
<tr>
<td>30</td>
<td>0.0105594</td>
<td>0.0915127</td>
</tr>
<tr>
<td>40</td>
<td>0.0726806</td>
<td>0.23843</td>
</tr>
<tr>
<td>50</td>
<td>0.254615</td>
<td>0.522479</td>
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<tr>
<td>60</td>
<td>0.28477</td>
<td>0.759431</td>
</tr>
</tbody>
</table>

Table 4.8. Parameters to evaluate the quality of the CBP of the 7:1 sample as functions of $\theta_{inc}$.

<table>
<thead>
<tr>
<th>$\theta_{inc}$</th>
<th>$H(\theta_{inc})$</th>
<th>$g_{avg}(\theta_{inc})$</th>
</tr>
</thead>
<tbody>
<tr>
<td>10</td>
<td>0.00160037</td>
<td>0.0249242</td>
</tr>
<tr>
<td>20</td>
<td>0.00178595</td>
<td>0.0416401</td>
</tr>
<tr>
<td>30</td>
<td>0.00818767</td>
<td>0.0949299</td>
</tr>
<tr>
<td>40</td>
<td>0.0586231</td>
<td>0.249451</td>
</tr>
<tr>
<td>50</td>
<td>0.160299</td>
<td>0.465593</td>
</tr>
<tr>
<td>60</td>
<td>0.17433</td>
<td>0.736478</td>
</tr>
</tbody>
</table>

Table 4.9. Parameters to evaluate the quality of the CBP of the 9:1 sample as functions of $\theta_{inc}$.

<table>
<thead>
<tr>
<th>$\theta_{inc}$</th>
<th>$H(\theta_{inc})$</th>
<th>$g_{avg}(\theta_{inc})$</th>
</tr>
</thead>
<tbody>
<tr>
<td>10</td>
<td>0.295102</td>
<td>0.138768</td>
</tr>
<tr>
<td>20</td>
<td>0.166418</td>
<td>0.101212</td>
</tr>
<tr>
<td>30</td>
<td>0.298627</td>
<td>0.351819</td>
</tr>
<tr>
<td>40</td>
<td>0.472629</td>
<td>0.620013</td>
</tr>
<tr>
<td>50</td>
<td>0.409609</td>
<td>0.903081</td>
</tr>
<tr>
<td>60</td>
<td>0.434223</td>
<td>1.35272</td>
</tr>
</tbody>
</table>

Table 4.10. Parameters to evaluate the quality of the CBP of the 1:1 sample as functions of $\theta_{inc}$.
Figure 4.21. $H(\theta_{inc})$ versus $\theta_{inc}$ for all samples.

Figure 4.22. $g_{avg}(\theta_{inc})$ versus $\theta_{inc}$ for all samples.
For any given sample, with some exceptions, there is an increase in $H(\theta_{\text{inc}})$ and $g_{\text{avg}}(\theta_{\text{inc}})$ with increasing $\theta_{\text{inc}}$. No pattern seems to be clear between samples when comparing $H(\theta_{\text{inc}})$ or $g_{\text{avg}}(\theta_{\text{inc}})$ for any given $\theta_{\text{inc}}$. This suggests that changing the ratio $h_1:h_2$, at least up until 9:1, does not significantly affect the strength or uniformity of discrimination between $R_{RR}$ and $R_{LL}$ within the circular Bragg regime.

### 4.1.3 Full-Width Half Maximum

The center wavelength $\lambda_{o}^{Br}(\theta_{\text{inc}})$ and the full-width half maximum (FWHM) $\Delta \lambda_{o}^{Br}(\theta_{\text{inc}})$ of the circular Bragg regime were also found from Figure 4.20 for $\theta_{\text{inc}} \in [10^\circ,60^\circ]$ every $10^\circ$. The value of $\lambda_{o}^{Br}$ is the wavelength of point 3 in Fig. 4.20. From point 3, the corresponding value of $R_{RR}$ (or $T_{RR}$) at point 4 was found. The values of $R_{RR}$ (or $T_{RR}$) at point 3 and 4 were averaged to find the half-maximum of the peak (or trough), (point 5 in Fig. 4.20). The value of $\Delta \lambda_{o}^{Br}$ was then measured at point 5. With the method just described, $\Delta \lambda_{o}^{Br}(\theta_{\text{inc}})$ was unobtainable at $\theta_{\text{inc}} = 70^\circ$ for some samples. Therefore, $\Delta \lambda_{o}^{Br}(\theta_{\text{inc}})$ was only found up to $\theta_{\text{inc}} = 60^\circ$.

The values of $\lambda_{o}^{Br}$ and $\Delta \lambda_{o}^{Br}$ from $R_{RR}$ and $T_{RR}$ were averaged and the functions

$$F(\theta_{\text{inc}}) = \frac{1}{\alpha} \frac{\lambda_{o}^{Br}(\theta_{\text{inc}})}{\cos^{1/2} \theta_{\text{inc}}}$$

(4.5)
\[ G(\theta_{\text{inc}}) = \frac{1}{\beta} \frac{\Delta \lambda_o^{Br}(\theta_{\text{inc}})}{\cos^{1/2} \theta_{\text{inc}}} \] (4.6)

were defined. Here, \( \alpha = \lambda_o^{Br}(0^\circ) \) and \( \beta = \Delta \lambda_o^{Br}(0^\circ) \), both taken from the \( T_{RR} \) spectrums. Figures 4.23 and 4.24 show the plots of \( F(\theta_{\text{inc}}) \) and \( G(\theta_{\text{inc}}) \) respectively. From Figure 4.23, it can be seen that all samples have a similar variation of \( \lambda_o^{Br} \) with respect to \( \theta_{\text{inc}} \). Figure 4.24 suggests there is no simple relationship between the FWHM values and the subdeposit-height ratio.
Figure 4.23. $F(\theta_{inc})$ versus $\theta_{inc}$ for all samples.

Figure 4.24. $G(\theta_{inc})$ versus $\theta_{inc}$ for all samples.
4.2 Criterion-Based Performance Parameters

If $R_{LL} \leq 0.05R_{RR}$ for satisfactory performance as a circular-polarization filter, then $f(\lambda_o, \theta_{inc}) \geq 1.8$. The enforcement of this criterion means that the circular Bragg regime fails to discriminate strongly enough for $\theta_{inc} > 40^\circ$ for all nine samples. With this criterion, the circular Bragg regimes used to calculate the data in Tables 4.2–4.10 were further refined for each $\theta_{inc}$. Tables 4.11–4.19 present $\lambda_o^{min}$, $\lambda_o^{max}$, and $\delta\lambda_o^{Br} = \lambda_o^{max} - \lambda_o^{min}$ as functions of $\theta_{inc} \in [10^\circ, 40^\circ]$ for all nine samples. The spectral width $\delta\lambda_o^{Br}$ of the circular Bragg regime narrows as $\theta_{inc}$ increases for every sample. In addition, there is a trend (with some exceptions) of smaller $\delta\lambda_o^{Br}$ as $h_1/h_2$ increases. In some instances, the criterion $f(\lambda_o, \theta_{inc}) \geq 1.8$ yielded a circular Bragg regime wider than shown in Figs. 4.1–4.18. In such cases, $\lambda_o^{min}$ and $\lambda_o^{max}$ were obtained directly by the process shown in Fig. 4.20.
### Table 4.11. Criterion-based parameters of the circular Bragg regime width of the 1:1 sample.

<table>
<thead>
<tr>
<th>$\theta_{inc}$</th>
<th>$\lambda_o^{min}$ (nm)</th>
<th>$\lambda_o^{max}$ (nm)</th>
<th>$\delta\lambda_o^{Br}$ (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>10</td>
<td>587.05</td>
<td>652.6</td>
<td>65.55</td>
</tr>
<tr>
<td>20</td>
<td>582.15</td>
<td>648.3</td>
<td>66.15</td>
</tr>
<tr>
<td>30</td>
<td>574</td>
<td>631</td>
<td>57</td>
</tr>
<tr>
<td>40</td>
<td>566</td>
<td>617</td>
<td>51</td>
</tr>
</tbody>
</table>

### Table 4.12. Criterion-based parameters of the circular Bragg regime width of the 1.5:1 sample.

<table>
<thead>
<tr>
<th>$\theta_{inc}$</th>
<th>$\lambda_o^{min}$ (nm)</th>
<th>$\lambda_o^{max}$ (nm)</th>
<th>$\delta\lambda_o^{Br}$ (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>10</td>
<td>596.2</td>
<td>664.2</td>
<td>68</td>
</tr>
<tr>
<td>20</td>
<td>594</td>
<td>656</td>
<td>62</td>
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<tr>
<td>30</td>
<td>585</td>
<td>643</td>
<td>58</td>
</tr>
<tr>
<td>40</td>
<td>575</td>
<td>628</td>
<td>53</td>
</tr>
</tbody>
</table>

### Table 4.13. Criterion-based parameters of the circular Bragg regime width of the 2:1 sample.

<table>
<thead>
<tr>
<th>$\theta_{inc}$</th>
<th>$\lambda_o^{min}$ (nm)</th>
<th>$\lambda_o^{max}$ (nm)</th>
<th>$\delta\lambda_o^{Br}$ (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>10</td>
<td>584.6</td>
<td>649.5</td>
<td>64.9</td>
</tr>
<tr>
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<td>580.9</td>
<td>642.8</td>
<td>61.9</td>
</tr>
<tr>
<td>30</td>
<td>576</td>
<td>631</td>
<td>55</td>
</tr>
<tr>
<td>40</td>
<td>571</td>
<td>616</td>
<td>45</td>
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</table>

### Table 4.14. Criterion-based parameters of the circular Bragg regime width of the 2.5:1 sample.

<table>
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<th>$\theta_{inc}$</th>
<th>$\lambda_o^{min}$ (nm)</th>
<th>$\lambda_o^{max}$ (nm)</th>
<th>$\delta\lambda_o^{Br}$ (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>10</td>
<td>598</td>
<td>663</td>
<td>65</td>
</tr>
<tr>
<td>20</td>
<td>595</td>
<td>656</td>
<td>61</td>
</tr>
<tr>
<td>30</td>
<td>588</td>
<td>643</td>
<td>55</td>
</tr>
<tr>
<td>40</td>
<td>582</td>
<td>628</td>
<td>46</td>
</tr>
</tbody>
</table>

60
<table>
<thead>
<tr>
<th>$\theta_{inc}$</th>
<th>$\lambda_o^{min}$ (nm)</th>
<th>$\lambda_o^{max}$ (nm)</th>
<th>$\delta \lambda_o^{Br}$ (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>10</td>
<td>603</td>
<td>667.3</td>
<td>64.3</td>
</tr>
<tr>
<td>20</td>
<td>598</td>
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<tr>
<td>40</td>
<td>585</td>
<td>634</td>
<td>49</td>
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</table>

**Table 4.15.** Criterion-based parameters of the circular Bragg regime width of the 3:1 sample.

<table>
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<tr>
<th>$\theta_{inc}$</th>
<th>$\lambda_o^{min}$ (nm)</th>
<th>$\lambda_o^{max}$ (nm)</th>
<th>$\delta \lambda_o^{Br}$ (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>10</td>
<td>609</td>
<td>668</td>
<td>59</td>
</tr>
<tr>
<td>20</td>
<td>602</td>
<td>661</td>
<td>59</td>
</tr>
<tr>
<td>30</td>
<td>595</td>
<td>650</td>
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<tr>
<td>40</td>
<td>589</td>
<td>633</td>
<td>44</td>
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</tbody>
</table>

**Table 4.16.** Criterion-based parameters of the circular Bragg regime width of the 5:1 sample.

<table>
<thead>
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<th>$\theta_{inc}$</th>
<th>$\lambda_o^{min}$ (nm)</th>
<th>$\lambda_o^{max}$ (nm)</th>
<th>$\delta \lambda_o^{Br}$ (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>10</td>
<td>609.1</td>
<td>667.9</td>
<td>58.2</td>
</tr>
<tr>
<td>20</td>
<td>603</td>
<td>660.55</td>
<td>57.55</td>
</tr>
<tr>
<td>30</td>
<td>595</td>
<td>646</td>
<td>51</td>
</tr>
<tr>
<td>40</td>
<td>590</td>
<td>629</td>
<td>39</td>
</tr>
</tbody>
</table>

**Table 4.17.** Criterion-based parameters of the circular Bragg regime width of the 7:1 sample.

<table>
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<tr>
<th>$\theta_{inc}$</th>
<th>$\lambda_o^{min}$ (nm)</th>
<th>$\lambda_o^{max}$ (nm)</th>
<th>$\delta \lambda_o^{Br}$ (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>10</td>
<td>615</td>
<td>672</td>
<td>54</td>
</tr>
<tr>
<td>20</td>
<td>609</td>
<td>668</td>
<td>59</td>
</tr>
<tr>
<td>30</td>
<td>601</td>
<td>651</td>
<td>50</td>
</tr>
<tr>
<td>40</td>
<td>596</td>
<td>627</td>
<td>31</td>
</tr>
</tbody>
</table>

**Table 4.18.** Criterion-based parameters of the circular Bragg regime width of the 9:1 sample.
<table>
<thead>
<tr>
<th>$\theta_{inc}$</th>
<th>$\lambda_o^{min}$ (nm)</th>
<th>$\lambda_o^{max}$ (nm)</th>
<th>$\delta \lambda^B_r$ (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>10</td>
<td>620.054</td>
<td>654.25</td>
<td>34.196</td>
</tr>
<tr>
<td>20</td>
<td>615.386</td>
<td>650.885</td>
<td>35.499</td>
</tr>
<tr>
<td>30</td>
<td>607.343</td>
<td>636.901</td>
<td>29.558</td>
</tr>
<tr>
<td>40</td>
<td>606.046</td>
<td>621.35</td>
<td>15.304</td>
</tr>
</tbody>
</table>

Table 4.19. Criterion-based parameters of the circular Bragg regime width of the finely ambichiral sample.

### 4.3 Cross-Section Morphology

Thickness measurements taken by the FESEM tool revealed that the samples ranged from 3.1 to 3.34 µm in thickness for reasons mentioned in Sec. 3.5. To get a clear image of morphology within the sample, away from any edge-growth effects, the samples were cleaved using the freeze-fracture technique. In addition, a CTF was grown and the angle $\chi$ measured, which was 37°. This is consistent with previous findings that columns tend to grow towards the normal with respect to $\chi_v$ [2].
Figure 4.25. Cross-sectional FESEM image of the 1:1 sample. The on-screen measurement tool reads 3.26 µm as the thickness.

Figure 4.26. Cross-sectional FESEM image of the 1.5:1 sample. The on-screen measurement tool reads 3.27 µm as the thickness.
Figure 4.27. Cross-sectional FESEM image of the 2:1 sample. The on-screen measurement tool read 3.1 µm as the thickness.

Figure 4.28. Cross-sectional FESEM image of the 2.5:1 sample. The on-screen measurement tool reads 3.26 µm as the thickness.
Figure 4.29. Cross-sectional FESEM image of the 3:1 sample. The on-screen measurement tool reads 3.34 µm as the thickness.

Figure 4.30. Cross-sectional FESEM image of the 5:1 sample. The on-screen measurement tool reads 3.34 µm as the thickness.
Figure 4.31. Cross-sectional FESEM image of the 7:1 sample. The on-screen measurement tool reads 3.22 µm as the thickness.

Figure 4.32. Cross-sectional FESEM image of the 9:1 sample. The on-screen measurement tool reads 3.20 µm as the thickness.
Figure 4.33. Cross-sectional FESEM image of the finely ambichiral sample. The on-screen measurement tool reads $3.25 \, \mu m$ as the thickness.

Figure 4.34. Cross-sectional FESEM image of the CTF sample. Nanocolumns grew at approximately $\chi = 37^\circ$. 

Helical nanocolumns are clearly seen in the 1:1 sample (Fig. 4.25), rising perpendicularly to the substrate. Even with a slight asymmetry, as in the 1.5:1 sample (Fig. 4.26), the nanocolumns look quite different compared to the 1:1 sample. Each nanocolumn takes on a C-shape that grows tilted locally, yet over 10 periods stays upright. The C-shape becomes better defined with increasing ratio $h_1/h_2$. As a thought experiment, one can imagine that for any ratio $h_1/h_2$ there will initially be tilting since one sub-deposit is shorter than the other. After one half of a period has been deposited, the longer sub-deposit will be on the other side making the structure lean the other way. Hence, after one period, the structure will overall stay perpendicular to the substrate. The 3:1 sample (Fig. 4.29), however, does seem to lean somewhat. To verify whether this was due to cleaving or some unaccounted factor during growth, a 5:1 sample was made. From the FESEM micrograph in Fig. 4.30, all helical nanocolumns look relatively straight, eliminating any previous doubts from the 3:1 sample. For the 5:1 sample, the deposition time for $h_2$ was calculated to be $\sim 0.905$ s, while the time to rotate $180^\circ$ and $180^\circ + \delta$ for each sub-deposit was on the order of $\sim 0.41$ s. Since the 5:1 sample still produced a strong CBP, I decided that the limit should be pushed even further. Therefore, samples with ratios of 7:1 (Fig. 4.31) and 9:1 (Fig. 4.32) were made. At this point, it was expected that a limit would be reached. Since the shorter sub-deposit becomes increasingly insignificant as $h_1/h_2$ is increased, at some point, the film essentially becomes a finely ambichiral STF with one sub-deposit, as in the method described.
in the first row of Fig. 1.6.

Therefore, an ambichiral STF was grown for comparison and is shown in Figure 4.33. Figures 4.31, 4.32, and 4.33, respectively, show that the morphologies of the 7:1, 9:1 and the finely ambichiral samples are indistinguishable. For the 7:1 and 9:1 samples, the deposition heights for the shorter subdeposit was $\sim 0.27\text{nm}$ and $\sim 0.21\text{ nm}$, respectively. These heights are comparable to the $\sim 0.16\text{ nm}$ deposited each turn. Even though $h_2$ did not seem to have an effect on growth, it is clearly still important for increasing the local linear birefringence as seen by the higher discrimination (i.e. $R_{RR} - R_{LL}$) in the circular Bragg regime when comparing Figs. 4.13–4.16 to Figs. 4.17 and 4.18.
Chapter 5

Conclusion

I set out to find how ASBD chiral STFs affect the CBP with light impinging at angle of incidence $\theta_{\text{inc}} \in [0^\circ, 70^\circ]$. The films were fabricated with ZnSe as the source material, due to its high index of refraction, optical transparency in the visible regime, and low-temperature vaporization at pressures below $10^{-4}$ Torr. Evaporation is easily achievable using resistive-heating PVD. Other benefits from this fabrication method include line-of-sight growth and a high as well as stable deposition rate.

Inside the deposition chamber, two motors were used to orient the substrate: one to ensure that the collimated vapor flux was directed at the chosen value of $\chi_v$ (= 20°) with respect to the substrate plane, and the other to rotate the substrate about its central normal axis. For all samples, the substrate was rotated by 180° between the first subdeposit of height $h_1$ and the second subdeposit $h_2$, and by 180° + $\delta$ between the subdeposit of height $h_2$ and the subdeposit of height $h_1$. 
The ratio of the two subdeposit heights $h_1/h_2$ was changed to create asymmetric growth, with $\delta = 3^\circ$ fixed. Chiral STFs were deposited for ratios $h_1:h_2$ of 1:1, 1.5:1, 2:1, 2.5:1, 3:1, 5:1, 7:1, and 9:1. Their optical characteristics were compared to those of a finely ambichiral STF (for which the ratio $h_1/h_2 \to \infty$). First, LCP light was incident and the LCP and RCP transmittances and reflectances were measured as functions of angle of incidence $\theta_{inc}$ and free-space wavelength $\lambda_o$. Then, RCP light was incident and the LCP and RCP transmittances and reflectances were similarly measured. Furthermore, the morphology of each sample was studied using FESEM images.

Red-shifting of the central wavelength $\lambda_{Br}^o$ of the circular Bragg regime for all $\theta_{inc} \in [0^\circ, 70^\circ]$ and narrowing of a criterion-based bandwidth $\delta \lambda_{Br}^o$ of the circular Bragg regime for all $\theta_{inc} \in [0^\circ, 40^\circ]$ with increasing $h_1/h_2$ were the two significant findings. If $R_{LL}$ was set to not exceed 5\% of $R_{RR}$, then all samples performed satisfactory only for $\theta_{inc} \leq 40^\circ$. Morphologically, the second subdeposit heights for samples 7:1 and 9:1 were so small that both of these samples were virtually indistinguishable from the finely ambichiral sample. The major conclusions of the research were as follows: chiral STFs grown using the ASBD method are more suitable than SBD chiral STFs for narrow-band circular polarization filters, ASBD chiral STFs are more promising as vertical-cavity-surface-emitting laser mirrors, and ASBD chiral STFs may be beneficial in surface multi-plasmonic resonance imaging.
5.1 Future Work

5.1.1 Narrow-band Circular Polarization Filter

This work has shown that the $\delta \lambda_o^{Br}$ can be reduced by 17% at $\theta_{inc} = 10^\circ$ and by 39% at $\theta_{inc} = 40^\circ$, if a 9:1 chiral STF is substituted by a 1:1 chiral STF. This would make chiral STFs grown using the ASBD method more suitable than SBD chiral STFs for narrow-band circular polarization filters. As mentioned in Sec. 4.1.1, the difference $R_{RR} - R_{LL}$ decreases slightly from the 7:1 sample to the 9:1 sample. The film with the best balance of narrow bandwidth and high $R_{RR} - R_{LL}$ is most likely between these two ratios.

5.1.2 Vertical-Cavity-Surface-Emitting Laser

One successful application of asymmetric chiral STFs of high $h_1/h_2$ ratio could be in circularly polarized lasers as cavity mirrors. The concept and realization of chiral-mirror-based micro-cavity light-emitting diodes and vertical-cavity-surface-emitting lasers has already been shown to work for symmetric chiral STFs [56], [57]. The center wavelength of the laser could be tuned by appropriately changing $h_1 + h_2 = h$ and/or by post-deposition annealing [58] and/or chemical etching [59].
5.1.3 Surface Multi-plasmonic Resonance Imaging

Surface multi-plasmonic resonance imaging (SMPRI) is an emerging technique to optically sense analytes in aqueous solutions [12]. A beam of light is incident upon one slanted face of a glass prism, on whose base a metal film and a chiral STF are successively deposited. The beam gets reflected at the base of the prism and emerges from the other slanted face of the prism to be captured by a charge-coupled device (CCD) spectrometer. There will be a range of angles of incidence for which surface-plasmon-polariton (SPP) waves will be excited to propagate along the interface of the metal and the chiral STF. When an SPP wave is excited, a sudden decrease in the light intensity measured by the spectrometer will occur. In order to sense analytes, in aqueous solution, the void network of the chiral STF can be infiltrated by the solution to change the dielectric properties of the chiral STF, and thus the angles of incidence for which a sudden decrease in the measured intensity occurs. Depositing the chiral STF using the SBD method can result in an increase in local linear birefringence, but a decreased void-network volume for the aqueous solution to penetrate. Use of a finely ambichiral STF allows for a larger void-network volume, but comes at the cost of decreased birefringence. Chiral STFs deposited using the ASBD method could provided a balance between these two characteristics.
5.1.4 Theoretical Model

Modeling can help pin-point interesting effects that may have been missed in this research because only a small number of ASBD chiral STFs were fabricated for experiments. Asymmetric growth is not represented by the rotation dyadic presented in Eqn. (2.5). Another model suggested by Venugopal [60] could be used to describe the asymmetric chiral structures. His model consists of creating a lattice with periodicity in two mutually orthogonal directions. At the center of each lattice point, a thin cylinder is placed. Venugopal’s model is described by the convolution of a cylinder function with two Dirac-delta functions, each controlling the periodicity in the $x$ and $y$ directions respectively. The lattice is then made to rotate as it rises in the $z$ direction with the functions, thus,

\[
x_a(z) = x_a(0) + R \sin \frac{\pi z}{\Omega}, \tag{5.1}
\]

\[
y_a(z) = y_a(0) + R \left(1 - \cos \frac{\pi z}{\Omega}\right), \tag{5.2}
\]

where $2\Omega$ is one period, $R$ is the helical sweep radius, and $x_a(0)$ and $y_a(0)$ are the positions of the centers of the cylinders in the plane $z = 0$. These equations develop the thin cylinders in a helical fashion as $z$ increases. For asymmetric growth, $x_a(z)$ and $y_a(z)$ need to be changed appropriately. The optical characteristics can be computed using rigorous coupled-wave analysis [60]. This model also has the added
benefit of accounting for the 3D periodicity of chiral STFs.
Reflectance data for the 1:1 sample when $\theta_{inc} = 10^\circ$ is shown to provide an example of how the raw optical intensity data was laid out in Microsoft Excel. Since the data spanned 3717 rows, only the beginning and the end are shown here to give an idea of how the meaningful data were picked out with Mathematica $^TM$ code.

```math
a = Import[
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ZnSe 300nm
1_1 (10_26_2015)\\RLL\\10.csv"];
Flatten[Table[a[[1 + (3717 - 52)i ;; 52 + (3717 - 52)i, 1 ;; 5]], {i, 0, 1}], 1]
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Appendix B

Mathematica Code for 2D-Density Plots

The code shown in this appendix was used for making the 2D-density plot of the 3:1 sample shown in Figs. 4.9 and 4.10. Similar code was used to create the 2D-density plots in Figs. 4.1-4.18. Subtracting out dark intensities from the remittance intensities resulted in some small negative values due to noise. This was fixed by imposing an ‘IF’ statement so that intensity values below zero were given a value of 0.
rmin = 1088;
rmax = 2641;

Darkdata1 = Import[
  "C:\Penn State\Research\PatM_ZnSe_ASBD\GOOD DATA\ZnSe 300nm 3_1 (11_5_2015)\Dark_Data_3_1.csv"
];

Darkdata2 = Darkdata1[[rmin ;; rmax, 3]];

ALLdata1 = Import[
  "C:\Penn State\Research\PatM_ZnSe_ASBD\GOOD DATA\ZnSe 300nm 3_1 (11_5_2015)\ALL_3_1.csv"
];

ALLdata = ALLdata1[[rmin ;; rmax, 3]];

ARRdata1 = Import[
  "C:\Penn State\Research\PatM_ZnSe_ASBD\GOOD DATA\ZnSe 300nm 3_1 (11_5_2015)\ARR_3_1.csv"
];

ARRdata = ARRdata1[[rmin ;; rmax, 3]];

λ1 = Import[
  "C:\Penn State\Research\PatM_ZnSe_ASBD\GOOD DATA\ZnSe 300nm 3_1 (11_5_2015)\Dark_Data_3_1.csv"
];

λ = λ1[[rmin ;; rmax, 2]];

θ = Flatten[{0, Table[i, {i, 10, 70, 2}]}];
RLLfiles = Table[
    Import["C:\Penn State\Research\PatM_ZnSe_ASBD\GOOD DATA\ZnSe 300nm 3_1 (11_5_2015)\RLL\" <> ToString[i] <> ".csv", "Data"], {i, 10, 70, 2}];

RLRfiles = Table[
    Import["C:\Penn State\Research\PatM_ZnSe_ASBD\GOOD DATA\ZnSe 300nm 3_1 (11_5_2015)\RLR\" <> ToString[i] <> ".csv", "Data"], {i, 10, 70, 2}];

RRLfiles = Table[
    Import["C:\Penn State\Research\PatM_ZnSe_ASBD\GOOD DATA\ZnSe 300nm 3_1 (11_5_2015)\RRL\" <> ToString[i] <> ".csv", "Data"], {i, 10, 70, 2}];

RRRfiles = Table[
    Import["C:\Penn State\Research\PatM_ZnSe_ASBD\GOOD DATA\ZnSe 300nm 3_1 (11_5_2015)\RRR\" <> ToString[i] <> ".csv", "Data"], {i, 10, 70, 2}];

TLLfiles = Table[
    Import["C:\Penn State\Research\PatM_ZnSe_ASBD\GOOD DATA\ZnSe 300nm 3_1 (11_5_2015)\TLL\0.csv"],
    Flatten[Table[Import["C:\Penn State\Research\PatM_ZnSe_ASBD\GOOD DATA\ZnSe 300nm 3_1 (11_5_2015)\TLL\" <> ToString[i] <> ".csv", "Data"], {i, 10, 70, 2}], 1];

TLRfiles = Table[
    Import["C:\Penn State\Research\PatM_ZnSe_ASBD\GOOD DATA\ZnSe 300nm 3_1 (11_5_2015)\TLR\0.csv"],
    Flatten[Table[Import["C:\Penn State\Research\PatM_ZnSe_ASBD\GOOD DATA\ZnSe 300nm 3_1 (11_5_2015)\TLR\" <> ToString[i] <> ".csv", "Data"], {i, 10, 70, 2}], 1];

TRLfiles = Table[
    Import["C:\Penn State\Research\PatM_ZnSe_ASBD\GOOD DATA\ZnSe 300nm 3_1 (11_5_2015)\TRL\0.csv"],
    Flatten[Table[Import["C:\Penn State\Research\PatM_ZnSe_ASBD\GOOD DATA\ZnSe 300nm 3_1 (11_5_2015)\TRL\" <> ToString[i] <> ".csv", "Data"], {i, 10, 70, 2}], 1];

TRRfiles = Table[
    Import["C:\Penn State\Research\PatM_ZnSe_ASBD\GOOD DATA\ZnSe 300nm 3_1 (11_5_2015)\TRR\0.csv"],
    Flatten[Table[Import["C:\Penn State\Research\PatM_ZnSe_ASBD\GOOD DATA\ZnSe 300nm 3_1 (11_5_2015)\TRR\" <> ToString[i] <> ".csv", "Data"], {i, 10, 70, 2}], 1];
Rll = Flatten[RLLfiles, 1];
Rlr = Flatten[RLRfiles, 1];
Rrl = Flatten[RRLfiles, 1];
Rrr = Flatten[RRRfiles, 1];

Tll = Flatten[TLLfiles, 1];
Tlr = Flatten[TLRfiles, 1];
Trl = Flatten[TRLfiles, 1];
Trr = Flatten[TRRfiles, 1];

Tll = Transpose[Table[Rll[[rmin + 3717 i ;; rmax + 3717 i, 3]], {i, 0, 30, 1}];
Tlr = Transpose[Table[Rlr[[rmin + 3717 i ;; rmax + 3717 i, 3]], {i, 0, 30, 1}];
RRL1 = Part[Table[1, {n, λ[[i]], θ}], Part[rlr, i]], {n, 1, 31, 1}], 1];

A = Table[If[Transpose[a][[i, n]] > 0, Transpose[a][[i, n]], 0], {i, 1, n, 1, 31}, 0, Transpose[a][[i, n]], 0],
               {i, 1, rmax - rmin + 1, 1}, {n, 1, 31, 1}];
B = ALLdata - Darkdata2;

rll = (A - B);

RLL1 = Flatten[Table[λ[[i]], θ[n + 1]], Part[Part[rll, i]], n],
               {i, 1, rmax - rmin + 1, 1}, {n, 1, 31, 1}], 1];

f = Table[If[Transpose[f][[i, n]] > 0, Transpose[f][[i, n]], 0],
               {i, 1, rmax - rmin + 1, 1}, {n, 1, 31, 1}];
G = ALLdata - Darkdata2;

rll = (F - G);

RLR1 = Flatten[Table[λ[[i]], θ[n + 1]], Part[Part[rlr, i]], n],
               {i, 1, rmax - rmin + 1, 1}, {n, 1, 31, 1}], 1];

h = Table[If[Transpose[h][[i, n]] > 0, Transpose[h][[i, n]], 0],
               {i, 1, rmax - rmin + 1, 1}, {n, 1, 31, 1}];
J = ARRdata - Darkdata2;

rll = (H - J);

RRL1 = Flatten[Table[λ[[i]], θ[n + 1]], Part[Part[rll, i]], n],
               {i, 1, rmax - rmin + 1, 1}, {n, 1, 31, 1}], 1];
k = Table[RRR[[All, i]] - Darkdata2, {i, 1, 31}];
K = Table[If[Transpose[k][[i, n]] > 0, Transpose[k][[i, n]], 0], {i, 1, rmax - rmin + 1}, {n, 1, 31}];
L = ARRdata - Darkdata2;
rrr = (K - L);
RRR1 = Flatten[Table[{λ[i], θ[n + 1], Part[Part[rrr, i], n]}, {i, 1, rmax - rmin + 1}, {n, 1, 31}]]
3_1_Density_Plots.nb
4 3
Printed by Wolfram Mathematica Student Edition
LL = ListDensityPlot[RLL1, PlotRange -> {All, All, All},
    BaseStyle -> {FontWeight -> Plain, FontFamily -> "Helvetica"},
    FrameStyle -> Directive[20, Plain], Frame -> True, FrameTicks ->
    {{10, 20, 30, 40, 50, 60, 70}, None}, {{500, 600, 700, 800, 900}, None}],
    FrameLabel -> {Style["\[\(\lambda_o\) (nm)\]", 30, Plain],
        Style["\[\(\theta\) (deg)\]", 30, Plain], Style["R L", 30, Plain]},
    ColorFunction -> "Rainbow", ColorFunctionScaling -> False];

LR = ListDensityPlot[RLR1, PlotRange -> {All, All, All},
    BaseStyle -> {FontWeight -> Plain, FontFamily -> "Helvetica"},
    FrameStyle -> Directive[20, Plain], Frame -> True, FrameTicks ->
    {{10, 20, 30, 40, 50, 60, 70}, None}, {{500, 600, 700, 800, 900}, None}],
    FrameLabel -> {Style["\[\(\lambda_o\) (nm)\]", 30, Plain],
        Style["\[\(\theta\) (deg)\]", 30, Plain], Style["R R", 30, Plain]},
    ColorFunction -> "Rainbow", ColorFunctionScaling -> False];

RL = ListDensityPlot[RRL1, PlotRange -> {All, All, All},
    BaseStyle -> {FontWeight -> Plain, FontFamily -> "Helvetica"},
    FrameStyle -> Directive[20, Plain], Frame -> True, FrameTicks ->
    {{10, 20, 30, 40, 50, 60, 70}, None}, {{500, 600, 700, 800, 900}, None}],
    FrameLabel -> {Style["\[\(\lambda_o\) (nm)\]", 30, Plain],
        Style["\[\(\theta\) (deg)\]", 30, Plain], Style["R L", 30, Plain]},
    ColorFunction -> "Rainbow", ColorFunctionScaling -> False];

RR = ListDensityPlot[RRR1, PlotRange -> {All, All, All},
    BaseStyle -> {FontWeight -> Plain, FontFamily -> "Helvetica"},
    FrameStyle -> Directive[20, Plain], Frame -> True, FrameTicks ->
    {{10, 20, 30, 40, 50, 60, 70}, None}, {{500, 600, 700, 800, 900}, None}],
    FrameLabel -> {Style["\[\(\lambda_o\) (nm)\]", 30, Plain],
        Style["\[\(\theta\) (deg)\]", 30, Plain], Style["R R", 30, Plain]},
    ColorFunction -> "Rainbow", ColorFunctionScaling -> False];

Legended[GraphicsGrid[{{LL, LR}, {RL, RR}}, ImageSize -> Scaled[.45]], BarLegend[
    {"Rainbow", {0, 1}}, LegendLayout -> "Column", LabelStyle -> {FontSize -> 20}]]
LL1 = ListDensityPlot[TLL1, PlotRange -> {All, All, All},
   BaseStyle -> {FontWeight -> Plain, FontFamily -> "Helvetica"},
   FrameStyle -> Directive[20, Plain], Frame -> True, FrameTicks ->
   {((0, 10, 20, 30, 40, 50, 60, 70), None), ((500, 600, 700, 800, 900), None)},
   FrameLabel -> {Style["λ_o (nm)", 30, Plain],
     Style["θ (deg)", 30, Plain], Style["T LL", 30, Plain]},
   ColorFunction -> "Rainbow", ColorFunctionScaling -> False];

LR1 = ListDensityPlot[TLR1, PlotRange -> {All, All, All},
   BaseStyle -> {FontWeight -> Plain, FontFamily -> "Helvetica"},
   FrameStyle -> Directive[20, Plain], Frame -> True, FrameTicks ->
   {((0, 10, 20, 30, 40, 50, 60, 70), None), ((500, 600, 700, 800, 900), None)},
   FrameLabel -> {Style["λ_o (nm)", 30, Plain],
     Style["θ (deg)", 30, Plain], Style["T LR", 30, Plain]},
   ColorFunction -> "Rainbow", ColorFunctionScaling -> False];

RL1 = ListDensityPlot[TRL1, PlotRange -> {All, All, All},
   BaseStyle -> {FontWeight -> Plain, FontFamily -> "Helvetica"},
   FrameStyle -> Directive[20, Plain], Frame -> True, FrameTicks ->
   {((0, 10, 20, 30, 40, 50, 60, 70), None), ((500, 600, 700, 800, 900), None)},
   FrameLabel -> {Style["λ_o (nm)", 30, Plain],
     Style["θ (deg)", 30, Plain], Style["T RL", 30, Plain]},
   ColorFunction -> "Rainbow", ColorFunctionScaling -> False];

RR1 = ListDensityPlot[TRR1, PlotRange -> {All, All, All},
   BaseStyle -> {FontWeight -> Plain, FontFamily -> "Helvetica"},
   FrameStyle -> Directive[20, Plain], Frame -> True, FrameTicks ->
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   FrameLabel -> {Style["λ_o (nm)", 30, Plain],
     Style["θ (deg)", 30, Plain], Style["T RR", 30, Plain]},
   ColorFunction -> "Rainbow", ColorFunctionScaling -> False];

Legended[GraphicsGrid[{LL1, LR1}, {RL1, RR1}], ImageSize -> Scaled[.45]], BarLegend[
  {"Rainbow", {0, 1}}, LegendLayout -> "Column", LabelStyle -> {FontSize -> 20}]]
Appendix C

Mathematica Code for Flatness Determination

This code was used to evaluate the right sides of Eqns. (4.2) and (4.4) of Sec. 4.1.2. This specific example is for the 9:1 sample. The matrix ‘u’ consists of the wavelengths outputted by the CCD spectrometer along with their $f(\lambda_o, \theta_{inc})$ values. Cases where $f(\lambda_o, \theta_{inc}) < 1.8$ were made to equal 0 so that the circular Bragg regime could easily be picked out by visual inspection.
In[16]:= rmin = 19;
   rmax = 3666;

   Darkdata1 = Import["C:\Penn State\Research\PatM_ZnSe_ASBD\GOOD DATA\ZnSe 300nm
   9_1 (1_6_2016)\Dark_Data_9_1.csv"];
   Darkdata2 = Darkdata1[[rmin ;; rmax, 3]];

   ALLdata1 = Import["C:\Penn State\Research\PatM_ZnSe_ASBD\GOOD DATA\ZnSe 300nm 9
   _1 (1_6_2016)\ALL_9_1.csv"];
   ALLdata = ALLdata1[[rmin ;; rmax, 3]];

   ARRdata1 = Import["C:\Penn State\Research\PatM_ZnSe_ASBD\GOOD DATA\ZnSe 300nm 9
   _1 (1_6_2016)\ARR_9_1.csv"];
   ARRdata = ARRdata1[[rmin ;; rmax, 3]];

   λ1 = Import["C:\Penn State\Research\PatM_ZnSe_ASBD\GOOD DATA\ZnSe 300nm 9
   _1 (1_6_2016)\Dark_Data_9_1.csv"];
   λ = λ1[[rmin ;; rmax, 2]];

   Δλ = Table[λ[[i]] - λ[[i + 1]], {i, 1, rmax - rmin, 1}];
RLLfiles = Table[
    Import["C:\Penn State\Research\PatM_ZnSe_ASBD\GOOD DATA\ZnSe 300nm 9_1 (1_6_2016)\RLL" <> ToString[i] <> ".csv", "Data"], {i, 10, 70, 2}];
RRRfiles = Table[Import["C:\Penn State\Research\PatM_ZnSe_ASBD\GOOD DATA\ZnSe 300nm 9_1 (1_6_2016)\RRR" <> ToString[i] <> ".csv", "Data"], {i, 10, 70, 2}];
Rll = Flatten[RLLfiles, 1];
Rrr = Flatten[RRRfiles, 1];
RLL = Transpose[Table[Rll[[rmin + 3717 i ;; rmax + 3717 i, 3]], {i, 0, 30, 1}]]; 
RRR = Transpose[Table[Rrr[[rmin + 3717 i ;; rmax + 3717 i, 3]], {i, 0, 30, 1}]]; 
a1 = Table[RLL[[All, i]] - Darkdata2, {i, 1, 31, 5}];
A1 = Table[If[Transpose[a1][[i, n]] <= 0, 0.00001, Transpose[a1][[i, n]]], {i, 1, rmax - rmin + 1, 1}, {n, 1, 7, 1}];
B1 = ALLdata - Darkdata2; 
R1 = Flatten[RLLfiles, 1];
Rrr = Flatten[RRRfiles, 1];
RLL = Transpose[Table[Rll[[rmin + 3717 i ;; rmax + 3717 i, 3]], {i, 0, 30, 1}]]; 
RRR = Transpose[Table[Rrr[[rmin + 3717 i ;; rmax + 3717 i, 3]], {i, 0, 30, 1}]]; 
a2 = Table[RRR[[All, i]] - Darkdata2, {i, 1, 31, 5}];
A2 = Table[If[Transpose[a2][[i, n]] <= 0, 0.00001, Transpose[a2][[i, n]]], {i, 1, rmax - rmin + 1, 1}, {n, 1, 7, 1}];
B2 = ARRdata - Darkdata2; 
r1 = Flatten[RLLfiles, 1];
rrr = Flatten[RRRfiles, 1];
z = -.5 (rrr - rll); 

z1 = Table[{λ[[i]], z[[i, 1]]}, {i, 1, Length[z1]}];
z2 = Table[{λ[[i]], z[[i, 2]]}, {i, 1, Length[z1]}];
z3 = Table[{λ[[i]], z[[i, 3]]}, {i, 1, Length[z1]}];
z4 = Table[{λ[[i]], z[[i, 4]]}, {i, 1, Length[z1]}];
z5 = Table[{λ[[i]], z[[i, 5]]}, {i, 1, Length[z1]}];
z6 = Table[{λ[[i]], z[[i, 6]]}, {i, 1, Length[z1]}];
z7 = Table[{λ[[i]], z[[i, 7]]}, {i, 1, Length[z1]}];
MatrixForm[z1];
y = Table[If[z1[[i, 2]] < 1.8, 0, z1[[i, 2]]], {i, 1, Length[z1]}]; 
MatrixForm[y];
u = Table[{λ[[i]], y[[i]]}, {i, 1, Length[z1]}]; 
MatrixForm[u];
\text{rmin}_1 = 1963; \\
\text{rmax}_1 = 2192; \\
\theta_{10} = \text{Transpose}[\{\lambda[[\text{rmin}_1 - 18 ;; \text{rmax}_1 - 18]], z[[\text{rmin}_1 - 18 ;; \text{rmax}_1 - 18, 1]]\}]; \\
\text{rmin}_2 = 1988; \\
\text{rmax}_2 = 2211; \\
\theta_{20} = \text{Transpose}[\{\lambda[[\text{rmin}_2 - 18 ;; \text{rmax}_2 - 18]], z[[\text{rmin}_2 - 18 ;; \text{rmax}_2 - 18, 2]]\}]; \\
\text{rmin}_3 = 2024; \\
\text{rmax}_3 = 2251; \\
\theta_{30} = \text{Transpose}[\{\lambda[[\text{rmin}_3 - 18 ;; \text{rmax}_3 - 18]], z[[\text{rmin}_3 - 18 ;; \text{rmax}_3 - 18, 3]]\}]; \\
\text{rmin}_4 = 2081; \\
\text{rmax}_4 = 2298; \\
\theta_{40} = \text{Transpose}[\{\lambda[[\text{rmin}_4 - 18 ;; \text{rmax}_4 - 18]], z[[\text{rmin}_4 - 18 ;; \text{rmax}_4 - 18, 4]]\}]; \\
\text{rmin}_5 = 2142; \\
\text{rmax}_5 = 2340; \\
\theta_{50} = \text{Transpose}[\{\lambda[[\text{rmin}_5 - 18 ;; \text{rmax}_5 - 18]], z[[\text{rmin}_5 - 18 ;; \text{rmax}_5 - 18, 5]]\}]; \\
\text{rmin}_6 = 2199; \\
\text{rmax}_6 = 2386; \\
\theta_{60} = \text{Transpose}[\{\lambda[[\text{rmin}_6 - 18 ;; \text{rmax}_6 - 18]], z[[\text{rmin}_6 - 18 ;; \text{rmax}_6 - 18, 6]]\}];

\text{avg}_{10} = \text{Sum}\left[\frac{\theta_{10}[n, 2]}{\text{Length}[\theta_{10}]}, \{n, 1, \text{Length}[\theta_{10}]\}\right]; \\
\text{avg}_{20} = \text{Sum}\left[\frac{\theta_{20}[n, 2]}{\text{Length}[\theta_{20}]}, \{n, 1, \text{Length}[\theta_{20}]\}\right]; \\
\text{avg}_{30} = \text{Sum}\left[\frac{\theta_{30}[n, 2]}{\text{Length}[\theta_{30}]}, \{n, 1, \text{Length}[\theta_{30}]\}\right]; \\
\text{avg}_{40} = \text{Sum}\left[\frac{\theta_{40}[n, 2]}{\text{Length}[\theta_{40}]}, \{n, 1, \text{Length}[\theta_{40}]\}\right]; \\
\text{avg}_{50} = \text{Sum}\left[\frac{\theta_{50}[n, 2]}{\text{Length}[\theta_{50}]}, \{n, 1, \text{Length}[\theta_{50}]\}\right]; \\
\text{avg}_{60} = \text{Sum}\left[\frac{\theta_{60}[n, 2]}{\text{Length}[\theta_{60}]}, \{n, 1, \text{Length}[\theta_{60}]\}\right];
\[
\begin{align*}
\text{flat10} &= \frac{1}{\lambda[[rmin1-18]] - \lambda[[rmax1-18]]} \\
&\times \left( \text{Sum}[\theta10[[i, 2]] - \text{avg10}^2 (\Delta\lambda[[i + rmin1 - rmin]]), \{i, 1, \text{Length}[\theta10], 1\}] \right)
\end{align*}
\]

\[
\begin{align*}
\text{flat20} &= \frac{1}{\lambda[[rmin2-18]] - \lambda[[rmax2-18]]} \\
&\times \left( \text{Sum}[\theta20[[i, 2]] - \text{avg20}^2 (\Delta\lambda[[i + rmin2 - rmin]]), \{i, 1, \text{Length}[\theta20], 1\}] \right)
\end{align*}
\]

\[
\begin{align*}
\text{flat30} &= \frac{1}{\lambda[[rmin3-18]] - \lambda[[rmax3-18]]} \\
&\times \left( \text{Sum}[\theta30[[i, 2]] - \text{avg30}^2 (\Delta\lambda[[i + rmin3 - rmin]]), \{i, 1, \text{Length}[\theta30], 1\}] \right)
\end{align*}
\]

\[
\begin{align*}
\text{flat40} &= \frac{1}{\lambda[[rmin4-18]] - \lambda[[rmax4-18]]} \\
&\times \left( \text{Sum}[\theta40[[i, 2]] - \text{avg40}^2 (\Delta\lambda[[i + rmin4 - rmin]]), \{i, 1, \text{Length}[\theta40], 1\}] \right)
\end{align*}
\]

\[
\begin{align*}
\text{flat50} &= \frac{1}{\lambda[[rmin5-18]] - \lambda[[rmax5-18]]} \\
&\times \left( \text{Sum}[\theta50[[i, 2]] - \text{avg50}^2 (\Delta\lambda[[i + rmin5 - rmin]]), \{i, 1, \text{Length}[\theta50], 1\}] \right)
\end{align*}
\]

\[
\begin{align*}
\text{flat60} &= \frac{1}{\lambda[[rmin6-18]] - \lambda[[rmax6-18]]} \\
&\times \left( \text{Sum}[\theta60[[i, 2]] - \text{avg60}^2 (\Delta\lambda[[i + rmin6 - rmin]]), \{i, 1, \text{Length}[\theta60], 1\}] \right)
\end{align*}
\]

\[
\begin{align*}
\text{Flat10} &= \text{Abs}[\text{avg10} - 2] \\
\text{Flat20} &= \text{Abs}[\text{avg20} - 2] \\
\text{Flat30} &= \text{Abs}[\text{avg30} - 2] \\
\text{Flat40} &= \text{Abs}[\text{avg40} - 2] \\
\text{Flat50} &= \text{Abs}[\text{avg50} - 2] \\
\text{Flat60} &= \text{Abs}[\text{avg60} - 2]
\end{align*}
\]
Appendix D

Mathematica Code for FWHM Plot

This code was used for the plots in Figs. 4.23 and 4.24. This specific example is for $T_{RR}$ of the 1:1 sample. The data were plotted and information about the wavelength and relative transmittance was gathered by right-clicking the plot and choosing 'Get Coordinates'. The same process was done for the $R_{RR}$ plot.
rmin = 1088;
rmax = 2641;

RRRdata = Import["C:\\Penn State\\Research\\PatM_ZnSe_ASBD\\GOOD DATA\\ZnSe 300nm 1_1 (10_26_2015)\\TRR\\0.csv"];
RRR = RRRdata[[rmin ;; rmax, 3]];

ARRdata = Import["C:\\Penn State\\Research\\PatM_ZnSe_ASBD\\GOOD DATA\\ZnSe 300nm 1_1 (10_26_2015)\\ARR.csv"];
ARR = ARRdata[[rmin ;; rmax, 3]];

Darkdata = Import["C:\\Penn State\\Research\\PatM_ZnSe_ASBD\\GOOD DATA\\ZnSe 300nm 1_1 (10_26_2015)\\Dark_Data_1_1.csv"];
Dark = Darkdata[[rmin ;; rmax, 3]];

λdata = Import["C:\\Penn State\\Research\\PatM_ZnSe_ASBD\\GOOD DATA\\ZnSe 300nm 1_1 (10_26_2015)\\ARR.csv"];

λ = λdata[[rmin ;; rmax, 2]];

a = RRR - Dark

A = Table[{λ[[i]], a[[i]]}, {i, 1, rmax - rmin + 1}];

ListPlot[A, {PlotRange -> All}]
Appendix E

Standard Operating Procedure

The following list outlines each deposition procedure:

1. MAKE SURE CURRENT CONTROLLER IS TURNED OFF. Insert tungsten boat between electrical contacts and tighten screws.

2. Add the source material to the tungsten boat, packing the material along the way until material is flush with sides of the boat.

3. Move the first motor to orient the substrate at the correct $\chi_v$.

4. Go through the following checklist: motor in position, shutter is closed, material packed and boat secured, QCM lifetime reads at least 83%, and correct deposition program has been selected.

5. Close chamber doors gently as to not knock any material from the boat.

Close the latches on the door.
6. Open high-vacuum valve and flip vacuum switch to ‘On’. Push on the chamber door to help create a seal. The high-vacuum pump will automatically turn on at the correct pressure.

7. Let the chamber sit for at least 3 hours, until the pressure reaches $\sim 2\epsilon(-6)$ Torr.

8. Turn on the current controller and then press ‘Voltage Out’, then ‘Fine’.

9. Increase the current at $\sim 0.1$ A/s.

10. Stop when power reaches about 190 W.

11. Open shutter, start second motor, and zero the thickness.

12. Once the thickness is reached, close shutter and ramp down current. Alternatively, if the motor has been programmed to stop automatically, close the shutter once the second motor has stopped.

13. Let chamber cool for thirty minutes.

14. Switch the high-vacuum to the ‘OFF’ position, close the high-vacuum valve, turn on the ‘VENT’ switch, and open the nitrogen gas tank.

15. Once the chamber is pressurized, close the nitrogen gas tank and turn off the ‘VENT’ switch.
Appendix F

Non-technical Abstract

Chiral structured thin films (STFs) are arrays of upright parallel nanohelixes that are fabricated by condensing a collimated vapor from a source material onto a rotating substrate, with the substrate normal tilted at an angle to the incoming vapor flux. Instead of continuous rotation, the substrate is rotated in small fractions of one revolution, which allows a significant amount of material to grow at a time in the direction of the incoming vapor flux. This is referred to as a subdeposit. The subdeposits form parallel nanohelixes. This morphology is due to a self-shadowing process, where the deposited material blocks other parts of the growing film from the incoming vapor flux, similar to how a tall building in a city casts a shadow, blocking the sun’s rays from hitting the streets. Another method involves making two subdeposits 180° apart before rotating back to the position of the first subdeposit and then introducing the small rotation. This method is called serial bideposition (SBD), wherein both subdeposits in SBD are of equal height.
If the subdeposits are of unequal heights, the deposition technique is known as asymmetric serial bideposition (ASBD).

Chiral STFs can be used in many applications. Because of the self-shadowing process described in the previous paragraph, there will be an interconnected space (void network) between the nanohelixes. This void network can be penetrated by solutions or gases that change the optical properties of the chiral STF. Thus, chiral STFs can be used for sensing capabilities.

One can also think of a nanohelix as a spring. Thus, a chiral STF is a bed of springs. In fact, it has been found chiral STFs possess an elastic displacement regime, and therefore can react to forces in the linear manner of Hooke’s law.

Before discussing the unique optical properties of chiral STFs, a brief background in optics is required. In 1865, James C. Maxwell demonstrated that the phenomenon we call ‘light’ is actually a propagating electromagnetic (EM) wave. An EM plane wave consists of an electric field oriented perpendicularly to a magnetic field, both of which are oscillating and both of which are perpendicular to the direction of propagation. For most applications, only the characteristics of the electric field are usually stated.

If the direction of oscillation of the electric field stays the same as the EM wave propagates, then the EM wave is referred to as linearly polarized. If the direction of the electric field does not stay in the same plane as the EM wave propagates, then the EM wave can either be elliptically or circularly polarized, the difference
between the two being that the first has an oscillating electric-field amplitude, and the second has a constant electric-field amplitude. In both cases, the electric field can rotate clockwise (left-handed) or counter-clockwise (right-handed) with respect to the propagation direction.

A chiral STF possesses a certain handedness (right or left i.e., counter-clockwise or clockwise) which can be determined by the sense of the substrate rotation during growth. Incoming circularly polarized light of the same handedness will get highly reflected from the chiral STF, while incoming circularly polarized light of the opposite handedness will not. Occurring in a limited portion of the spectral regime, this circular-polarization-discriminatory phenomenon is called the circular Bragg phenomenon. These characteristics make chiral STFs useful as circular-polarization filters.

This work focused on the circular Bragg phenomenon exhibited by ASBD chiral STFs. Films were made with various subdeposit-height ratios. Left-handed and right-handed circularly polarized light were incident upon all the samples for varying angles of incidence. The spectra of the measured reflectances and transmittances show that ASBD chiral STFs can function as narrow-band circular-polarization filters. Applications of ASBD chiral STFs may possibly be extended to resonance cavity mirrors in vertical-cavity-surface-emitting lasers and for sensing purposes with surface multi-plasmonic resonance imaging.
Bibliography


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