Polarization-selective diffraction for display applications

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Polarization-Selective Diffraction for Display Applications

PROEFSCHRIFT

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door

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Summary

Polarizers are commonly used optical elements in devices such as liquid crystal displays (LCDs) to create polarized light. LCDs need linear polarized light to be able to switch it. Linear polarized light is created by absorbing the light of the orthogonal linear polarization direction. Therefore, about 50% of the light is lost. To produce color displays, absorbing red, green and blue color filters are used, each absorbing about 66% of the light. Both absorbing elements contribute to a very inefficient use of light in LCDs. Overall only about 5% of the light, which is generated, is transmitted in a liquid crystal display (LCD). Especially for mobile devices with an LCD, like laptops and mobile phones, this inefficient use of light has a negative impact on the battery-life.

In this thesis an alternative and more efficient non-absorbing polarizer will be presented, which is also capable of separating colors. It will be shown that holographic slanted phase gratings are able to diffract polarization-selective from a waveguide without absorption. Linear polarized light is coupled out by diffraction towards the viewer, while the orthogonal polarization direction remains waveguided. As the diffraction is wavelength-selective, colors are automatically spatially separated and directed towards the corresponding color-pixel. This can eventually lead to a very efficient display.

Two types of gratings are experimentally investigated, which are capable of diffracting in a polarization-selective and collimated way without absorption. First, a novel nano-porous material is developed, which achieves a high index modulation as a volume hologram, while maintaining a relatively easy fabrication. Requirements, to couple out polarization-selective waveguided light by diffraction, are met based on theoretical predictions of Kogelnik’s coupled wave theory and experimentally verified. Polarization contrasts of 7.5, 35 and 11.2 were measured for red, green and blue colors. Second, a slanted polymer dispersed liquid crystal grating is developed to implement polarization-selective diffraction (out-coupling) of light trapped within a waveguide. The polarization-selectivity in this case occurs on the basis of matching the ordinary refractive index and mismatching the extraordinary refractive index of aligned liquid crystals with the refractive index of the isotropic polymer. Primary findings include polarization contrasts of 10, unidirectionality of 18, and collimation of each color within ±3° for the whole range of visible wavelengths.

The refractive index profiles of nano-porous gratings have been modeled and fitted to the diffraction efficiency calculated with the rigorous coupled wave theory. The angular diffraction efficiency of various diffracted orders has been used to determine the refractive index amplitude and shape. Only the amplitude seems to be of importance on the diffraction of thick gratings. For thin gratings the shape of the refractive index profile determines the relative intensity of the diffracted orders. The diffraction efficiency of gratings, that are in between both thickness regimes, is determined by the refractive index profile and amplitude of the refractive index. A block shaped grating profile with a fill factor is a more accurate
method to model the diffraction efficiencies of nano-porous gratings than a sinusoidal shaped profile.

Polarization-selective diffraction can also be achieved with polarization holograms. In this work a theoretical framework is presented to describe the formation and diffraction of these holograms. Most existing theoretical models describe the diffraction and formation of only linear birefringent modulating polarization gratings. In this case, general polarization holograms are described using modulating Stokes parameters combined with linear and circular birefringence. With this theoretical framework a polarization grating can be designed to diffract any state of polarization selectively with a high diffraction efficiency by choosing an interference pattern and a polarization sensitive recording material. To illustrate the applicability of the theoretical framework, several examples are presented, revealing the polarization-selectivity of diffraction of polarization holograms. A classification of polarization holograms is made based on the material responses and recorded interference patterns.

Out-coupling structures that improve the efficiency of display systems, are not limited to LCD applications. They can also be applied to organic light-emitting diode (OLED) based displays. Although polarization-selective emission is not required for OLED displays, gratings can still be used to improve their efficiency. Light is trapped inside the OLED due to total internal reflection. Coupling this light out increases the light emission and therefore the efficiency. Proof of principle is given, that functional OLEDs can be made from structured PEDOT and PPV layers by pulsed holography. Potentially, these gratings can result in more efficient OLEDs with a controlled angular emission. Also gratings have been made in the ITO-layer of an OLED. Different types of ITO were structured successfully with different kind of diffraction patterns.

In conclusion, it is shown that properly tuned diffraction gratings can be used to increase the efficiency of both LCD and LED displays. An excellent agreement is observed between theoretical predictions and practical results for slanted nano-porous and polymer dispersed liquid crystal gratings to diffract waveguided light polarization-selectively. The polarization gratings are useful to be able to diffract light of any state of polarization efficiently.
### List of symbols

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Definition</th>
</tr>
</thead>
<tbody>
<tr>
<td>$a$</td>
<td>Minor axis of the ellipse</td>
</tr>
<tr>
<td>$A$</td>
<td>Amplitude of the electric field</td>
</tr>
<tr>
<td>$A_{\parallel}$</td>
<td>Absorbance with a polarization parallel to the excitation</td>
</tr>
<tr>
<td>$A_{\perp}$</td>
<td>Absorbance with a polarization perpendicular to the excitation</td>
</tr>
<tr>
<td>$b$</td>
<td>Major axis of the ellipse</td>
</tr>
<tr>
<td>$c$</td>
<td>Speed of light</td>
</tr>
<tr>
<td>$C$</td>
<td>Aperture constant</td>
</tr>
<tr>
<td>$c_R$</td>
<td>Obliquity factor of reference wave</td>
</tr>
<tr>
<td>$c_S$</td>
<td>Obliquity factor of source wave</td>
</tr>
<tr>
<td>$d$</td>
<td>Thickness of a layer or grating</td>
</tr>
<tr>
<td>$D_A$</td>
<td>Diffraction amplitude</td>
</tr>
<tr>
<td>$D_m$</td>
<td>Far field amplitude of diffraction order $m$</td>
</tr>
<tr>
<td>$E$</td>
<td>Electric field vector</td>
</tr>
<tr>
<td>$E_0$</td>
<td>Plane wave amplitude-polarization vector</td>
</tr>
<tr>
<td>$E_I$</td>
<td>The total electric field ‘above’ a grating</td>
</tr>
<tr>
<td>$E_{\text{holo}}$</td>
<td>The electric field of an hologram</td>
</tr>
<tr>
<td>$E_{II}$</td>
<td>The total electric field ‘below’ a grating</td>
</tr>
<tr>
<td>$E_G$</td>
<td>The total electric field inside a grating</td>
</tr>
<tr>
<td>$E_{in}$</td>
<td>The electric field towards a grating</td>
</tr>
<tr>
<td>$E_{m}$</td>
<td>Electric field of plane-wave $m$</td>
</tr>
<tr>
<td>$E_n$</td>
<td>Energy level of an atom</td>
</tr>
<tr>
<td>$E_{\text{tot}}$</td>
<td>Total the electric field</td>
</tr>
<tr>
<td>$E_x$</td>
<td>X-component of the electric field</td>
</tr>
<tr>
<td>$E_y$</td>
<td>Y-component of the electric field</td>
</tr>
<tr>
<td>$F$</td>
<td>Fresnel number</td>
</tr>
<tr>
<td>$h$</td>
<td>Planck’s constant</td>
</tr>
<tr>
<td>$H_I$</td>
<td>The total magnetic field ‘above’ a grating</td>
</tr>
<tr>
<td>$H_{II}$</td>
<td>The total magnetic field ‘below’ a grating</td>
</tr>
<tr>
<td>$H_G$</td>
<td>The total magnetic field inside a grating</td>
</tr>
<tr>
<td>$I$</td>
<td>Intensity of light</td>
</tr>
<tr>
<td>$J$</td>
<td>Jones vector</td>
</tr>
<tr>
<td>$J_p$</td>
<td>P-component of the Jones vector</td>
</tr>
<tr>
<td>$J_s$</td>
<td>S-component of the Jones vector</td>
</tr>
<tr>
<td>$k$</td>
<td>Wave vector</td>
</tr>
<tr>
<td>$k_{\parallel}$</td>
<td>X-component of a wave vector of a diffraction order</td>
</tr>
<tr>
<td>$k_{\perp}$</td>
<td>Z-component of a wave vector of a diffraction order ‘above’ the grating</td>
</tr>
<tr>
<td>$k_{II,\parallel}$</td>
<td>Z-component of a wave vector of a diffraction order ‘below’ the grating</td>
</tr>
<tr>
<td>$L$</td>
<td>Cavity length of a laser</td>
</tr>
<tr>
<td>$L_c$</td>
<td>Vector in Stokes space pointing to $C_c$</td>
</tr>
<tr>
<td>$l_c$</td>
<td>Spectral line-width of the laser</td>
</tr>
<tr>
<td>$N_n$</td>
<td>Population of an energy level of an atom</td>
</tr>
<tr>
<td>$n$</td>
<td>Refractive index</td>
</tr>
<tr>
<td>$n_0$</td>
<td>Average refractive index</td>
</tr>
<tr>
<td>$n_1$</td>
<td>Refractive index modulation amplitude</td>
</tr>
</tbody>
</table>
\( \Delta n \) - Birefringence amplitude
\( \Delta n_i \) - Induced birefringence
\( \Delta n_{lin} \) - Linear birefringence amplitude \( (n_e - n_o) \)
\( \Delta n_{cir} \) - Circular birefringence amplitude \( (n_r - n_l) \)
\( N \) - Refractive index matrix
\( N_{bir} \) - Birefringence amplitude
\( N_{lin} \) - Linear birefringence amplitude
\( N_{cir} \) - Circular birefringence amplitude
\( n_\| \) - Refractive index parallel to the polarization of the excitation
\( n_{\perp} \) - Refractive index perpendicular to the polarization of the excitation
\( n_e \) - Refractive index for left-handed circular polarized light
\( n_o \) - Ordinary refractive index of a liquid crystal
\( n_r \) - Refractive index for right-handed circular polarized light
\( P \) - Pitch of a helix
\( Q \) - Distinctive factor between thick and thin gratings
\( r \) - Coordinate
\( R \) - Distance of the aperture to screen
\( R_0 \) - Electric field of a reflected order
\( S_0 \) - Stokes parameter describing intensity
\( S_1 \) - Stokes parameter describing linear polarized light
\( S_2 \) - Stokes parameter describing linear polarized light
\( S_3 \) - Stokes parameter describing circular polarized light
\( s_0 \) - Stokes parameter describing birefringence magnitude
\( s_1 \) - Stokes parameter describing linear birefringence
\( s_2 \) - Stokes parameter describing linear birefringence
\( s_3 \) - Stokes parameter describing circular birefringence
\( t \) - Time
\( T \) - Transfer matrix
\( T_0 \) - Isotropic refractive index amplitude
\( T_1 \) - Electric field of a transmitted order
\( \alpha_S \) - Order parameter
\( \delta \) - Phase
\( \epsilon \) - Dielectric permittivity
\( \eta \) - Diffraction efficiency
\( \eta_i \) - Diffraction efficiency of diffracted order \( i \)
\( \theta \) - Polar angle
\( \theta_B \) - Bragg-angle
\( \kappa_\| \) - Photo-anisotropy parallel to the polarization direction
\( \kappa_{\perp} \) - Photo-anisotropy perpendicular to the polarization direction
\( \lambda \) - Wavelength of light
\( \lambda_B \) - Bragg-wavelength
\( \Lambda \) - Periodicity of grating
\( \mu \) - Magnetic permeability

\( C^2 / Jm \) - Dielectric permittivity
\( m \) - Wavelength of light
\( m \) - Magnetic permeability
<table>
<thead>
<tr>
<th>Symbol</th>
<th>Unit</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\nu$</td>
<td>Hz</td>
<td>Frequency of radiated photons</td>
</tr>
<tr>
<td>$\nu_m$</td>
<td>Hz</td>
<td>Emission frequency of the laser</td>
</tr>
<tr>
<td>$\nu_s$</td>
<td>-</td>
<td>Modulation parameter of s-polarized light</td>
</tr>
<tr>
<td>$\nu_p$</td>
<td>-</td>
<td>Modulation parameter of p-polarized light</td>
</tr>
<tr>
<td>$\xi$</td>
<td>-</td>
<td>Dephasing parameter</td>
</tr>
<tr>
<td>$\rho$</td>
<td>-</td>
<td>Orientation of the linear birefringence</td>
</tr>
<tr>
<td>$\upsilon$</td>
<td>-</td>
<td>Measure of dephasing</td>
</tr>
<tr>
<td>$\phi$</td>
<td>rad</td>
<td>Azimuthal angle</td>
</tr>
<tr>
<td>$\varphi_G$</td>
<td>rad</td>
<td>Slant angle of a grating</td>
</tr>
<tr>
<td>$\chi$</td>
<td>rad</td>
<td>Ellipticity angle</td>
</tr>
<tr>
<td>$\chi_e$</td>
<td>-</td>
<td>Eigen-polarization state</td>
</tr>
<tr>
<td>$\psi$</td>
<td>rad</td>
<td>Polarization angle</td>
</tr>
<tr>
<td>$\Psi$</td>
<td>rad</td>
<td>Angle of measurement from a backlight system</td>
</tr>
</tbody>
</table>
Chapter 1

Introduction

The performance of devices such as flat panel displays, optical recording media and illumination systems depends critically on the control of the direction, polarization, and degree of collimation for the entire range of visible wavelengths. Consider, for a moment, the operation of the dominant application involving all three aspects: liquid crystal displays (LCDs). LCDs are thin, flat displays consisting of a stack of layers, which are able to address a number of color or monochrome pixels, arrayed in front of a light source or reflector. The working principle of a LCD is to utilize liquid crystals as switches to control the amount of passed light.

To be able to switch the light a stack comprising of a nematic liquid crystal layer sandwiched between two glass plates is used. The glass is covered with electrodes, typically transparent Indium Tin Oxide (ITO), and is coated with a rubbed polyimide orientation layer. The orientation layer aligns the nematic liquid crystals uniformly parallel to the rubbed direction. Figure 1.1 illustrates a common configuration of such a stack: the rubbing directions of the substrates are perpendicular, the liquid crystal twists 90 degrees over the thickness of the layer and at the bottom and top of the display linear absorbing polarizers are placed with an perpendicular (crossed) orientation.

When light is transmitted through the first polarizer, it becomes linear polarized, rotates 90 degrees due to the liquid crystal layer before passing through the linear polarizer at the front. This is called the bright state of an LCD. Applying a voltage on the liquid crystal layer orients the liquid crystals along the electric field and removes its rotation. In this state the linear polarized light does not rotate anymore and is absorbed by the second linear polarizer in the front of the display. This is called the dark state of the LCD. Applying intermediate voltages creates grey states. Adding color filters (red, green and blue) makes color images possible.

Many variations on the illustrated stack exist to serve the needs of the wide spectrum of LCD applications. The most significant aspects are the lighting system and the switching method. LCDs can be either transmissive, reflective or transflective, depending on the location of the light source. A transmissive LCD is illuminated from the back by a backlight and light is transmitted through the stack. This type of LCDs have applications that require high luminance levels such as computer displays, televisions, personal digital assistants, and mobile phones. Reflective LCDs, often found in digital watches, calculators and phones, are illuminated by external light using a (diffusing) reflector behind the display. The absence of an active light source significantly reduces power consumption, making this type of display...
suitable for devices requiring a low power consumption. Transflective LCDs make use of a semi-transparent mirror and a backlight. They work either in transmissive mode with backlight, when there is not enough environment light, or in reflective mode, when there is enough environment light.

When we focus on different switching methods we can discriminate Twisted Nematic (TN), In-Plane Switching (IPS), Multi-domain Vertical Alignment (MVA), and Patterned Vertical Alignment (PVA). The TN display is the most common consumer display type, due to its relative simple design and addressing scheme, resulting in a relative low price [1]. This switching method is applied in devices which do not require high switching speeds and large viewing angles. IPS, developed by Hitachi [2], utilizes a single-sided integrated electrode structure improving viewing angles and therefore color reproduction. Initially this improvement was traded for response time, which was initially on the order of 50 ms. Currently, improvements on this switching method are available that overcome the response time limitation. IPS technology commonly appear in the high end display market. MVA, originally developed by Fujitsu [3], is a compromise between TN and IPS. It is based on the less common liquid crystal with a negative dielectric anisotropy and it achieves fast pixel response times, has a wide viewing angle, and achieves a high contrast at the cost of brightness. MVA targets the same display market as IPS. PVA is a recent improvement on the MVA technology developed by Samsung [4] and it has an improved contrast ratio and response times.

Despite the wide variety of LCDs, light management in all of them is rather inefficient. For transmissive displays only a few percent of the generated light passes through the display in the bright state (see Figure 1.1). The main reasons for this inefficiency will be elaborated further. The light coming from a cold cathode fluorescent lamp (CCFL) enters a waveguide. Light couples out of the waveguide by uniform scattering of printed scattering elements with an efficiency of about 80%. Additionally, a lot of light is lost due to the first linear polarizer absorbing more than 50% of the light. Furthermore, color filters absorb more than 66% of the light (since each color filter only transmits about 33% of the spectrum). This, combined
Introduction

Figure 1.2: A slanted grating on a waveguide indicating the wavelength dependence of the diffracted out coupling angle.

with some other losses due to the reflection and absorption, results in the total light efficiency of a transmissive display of only about 5%.

The current method to improve the light efficiency of the display is by using a brightness enhancement film (as patented by 3M under the commercial name Vikuiti\textsuperscript{TM}). The brightness enhancement film utilizes refraction and reflection to increase the efficiency of your backlight. It refracts light within the viewing cone toward the viewer. Light outside this angle is reflected back and recycled until it exits at the proper angle. The film provides a 37% increase in on-axis brightness.

The on-axis brightness can be increased by up to 68% by using an additional dual brightness enhancement film (also from 3M). Instead of absorbing 50% of the light, by a linear absorbing polarizer, inserting a reflective polarizer (Vikuiti\textsuperscript{TM} DBEF film) between the bottom polarizer and the light source prevents this light from being absorbed and recycles it back into the display.

In this thesis an alternative design for an out-coupling structure on a waveguide is presented, making the use of absorbing polarizers and color filters partly or wholly redundant. As previously reported [5], waveguided light is diffracted (coupled out) towards the viewer more efficiently by a slanted grating as indicated in Figure 1.2. Display applications have several constraints and requirements with respect to light out-coupling, such as:

- Light has to be coupled out at near normal angles to the waveguide. Since the angle of diffraction is wavelength dependent, ’green’ (550 nm) is chosen to be exactly at the normal.

- Production of the out-coupling structure should be feasible on a typical display size.

- The materials of the out-coupling structure need to be transparent and clear for visible wavelengths.

- Preferably, light that is coupled out needs to be linear polarized. Other polarization states need to remain in the waveguide to be recycled.

Additionally it is desired to separate the colors. Currently, displays use absorbing color filters to produce red, green and blue light, implying that 66% of the light is lost. When different colors are coupled out under different angles, they can be directed to the corresponding color pixel, avoiding the absorption and thus improving the efficiency even further, as indicated in Figure 1.2.
As shown by Jagt et al. [5], a slanted holographic diffraction grating can meet the above requirements, provided that it is designed carefully with respect to refractive index modulation, layer thickness, periodicity and slant angle. In this thesis these polarization-selective slanted gratings are demonstrated. It will be shown how to predict their polarization-selectivity, how to produce these gratings and how they diffract. The gratings are made using holography, as it is an easy and flexible tool to make gratings with a wide variety of properties.

Out-coupling gratings, that improve the efficiency of display systems, are not only applicable to LCD applications. They can also be applied to organic light-emitting diode (OLED) based displays. Although polarization-selective emission is not required for OLED displays, gratings can still be used to improve their efficiency. A lot of light is trapped inside the OLED due to total internal reflection, coupling this light out increases the total amount of emitted light and therefore the efficiency.

1.1 Holography

In 1948 Dennis Gabor, a scientist at Imperial College London, reasoned he could record not only the brightness but also the intensity variation by interference between light shining through an object and standard reference light. This discovery triggered new photographic techniques that we now know as holography. Using a mercury arc lamp with a green filter, Gabor produced the first "in-line" transmission holograms in a thin transparency. In essence the process of creating holographic patterns consists of three steps:

- creating an interference pattern of coherent polarized light,
- capturing the interference pattern in a photo-sensitive film,
- reconstructing the hologram by diffraction of light from this film.

Despite the fact that Gabor hinted that spectrally pure (coherent) light would provide further optimization, holography lay dormant until 1960 when coherent light sources became available with the invention of the laser by T.A. Maiman [6]. Another major step in holography is contributed to Stephen A. Benton, who invented white-light transmission holography in 1968 during his research activities in holographic television at Polaroid Research Laboratories. These type of holograms can be viewed in regular "white" light and are visible as a "rainbow" image. Benton’s invention is particularly significant as it enabled mass production of holograms using an embossing technique. These holograms are "printed" by stamping the interference pattern into polymeric materials. The resulting hologram can be duplicated millions of times at extremely low costs. Consequently, embossed holograms are now being used in a wide spectrum of applications including publishing, advertising, and security. Commonly, 3D holographic images are applied in security features of credit cards, bank notes, and important documents.

The construction of these type of holograms typically involves the use of a photographic plate, which is illuminated by a coherent laser. The laser is first split into a reference beam, which goes unchanged to the photographic plate, and an object beam. The object beam hits the photographic plate after being reflected from an object as shown in Figure 1.3. In this case a mirror is used as a reflecting object, similar to the reference beam. This results in a grating (a regular pattern).
Two recording techniques are discriminated, depending on the orientation of the recording film with respect to the beams. If the object beam is reconstructed by reflection, the technique is referred to as reflection holography (Figure 1.3b). Reflection holograms have the grating direction in the direction along the layer-thickness. On the other hand, if the photographic film is transparent and the object beam is diffracted from the photographic plate, the technique is referred to as transmission holography (Figure 1.3a). Transmission holograms have the grating in a direction in the film.

Holographic films currently are obtained by recording (writing) holographic information in the form of the characteristic interfering light patterns in a photo-sensitive material. The photosensitive material normally is applied as a thin layer on a transparent or black substrate. In this thesis the formation and diffractive properties of several holographic films will be described. In its broadest perspective, the objective of this thesis is to create optical components based on holographic polymer films that give not only control over the direction and amount of light that is diffracted, but also its state of polarization. Basically, light is used to form patterns in polymer films with the purpose to control light after the polymer films have been formed. As will be shown, holography is an excellent tool to create such films by carefully selecting the polarization states, wavelength and angle of incidence of the interfering light in combination with light sensitive (polymer) materials and films.

Potential applications of these holographic films are envisioned in the field of flat-panel displays with possible extensions to sensor, telecommunication, and storage applications, which work on the principle of switching linear polarized light. As explained before, current technologies use absorbing polarizers and scattering out-coupling structures, which make them very inefficient because of the loss of light due to absorption and uncontrolled scattering. Holographic films, capable of diffracting light polarization-selectively, could replace the absorbing polarizers making the flat panel displays much more efficient.

In order to appreciate all the relevant aspects of holographic films, several concepts need to be elaborated first. The following sections discuss methods to describe light, the generation of coherent light, holographic material properties and the application of holography.
1.2 Polarization of light

Light is an electromagnetic transverse wave, consequently it is described by the direction of an oscillating electric field in the plane perpendicular to the direction of propagation. This oscillation direction is called polarization. The electric field vector \( \mathbf{E}(\mathbf{r}) \), depends on the spatial position \( \mathbf{r} \) of a coherent laser. The electrical field of monochromatic polarized light traveling in the \( z \)-direction can be described in good approximation by monochromatic sinusoidal plane-wave:

\[
\mathbf{E}(\mathbf{r}, t) = \begin{pmatrix}
E_x \cos(kz - \omega t + \alpha_x) \\
E_y \cos(kz - \omega t + \alpha_y) \\
0
\end{pmatrix},
\]

with \( \omega \) the angular frequency of the wave, \( \alpha_x \) and \( \alpha_y \) the phases and

\[
E_x = |\mathbf{E}| \cos \psi \quad E_y = |\mathbf{E}| \sin \psi,
\]

with

\[
\psi = \arctan \left( \frac{E_y}{E_x} \right),
\]

and

\[
|\mathbf{E}|^2 = E_x^2 + E_y^2.
\]

For plane waves the electric field vector can be divided into two components perpendicular to its propagation direction. Denoting these components as \( E_x \) and \( E_y \) with \( z \) the propagation direction results in the diagrams indicated in Figure 1.5a-c. The \( E_x \) and \( E_y \) components have the same frequency, but can differ in amplitude and phase, resulting in three different classes.

**Linear polarization** If the \( E_x \) and \( E_y \) component are in phase or in anti-phase the polarization is called linear as indicated in Figure 1.5a. As can be seen, the projection of the wave on the \((x, y)\)-plane forms a line in this case. The direction of this line depends on the relative amplitude of the \( E_x \) and \( E_y \) components. The ratio of the two components is a constant over time.

**Circular polarization** Circular polarization occurs when the two orthogonal components are ninety degrees out of phase and have the same amplitude. In this case, one component is zero when the other component is at maximum or minimum amplitude as illustrated in Figure 1.5b. There are two possible phase relationships that satisfy this requirement: \( E_x \) can be leading ninety degrees or lagging ninety degrees compared to \( E_y \) resulting in right-hand circular polarization or left-hand circular polarization, respectively.
Fig. 1.5: Different polarization states of light resulting from phase differences of $E_x$ and $E_y$. The wave is propagating in the $+z$-direction.

**Elliptical polarization** All other cases, i.e. when the phases are not a multiple of $\pi/2$ and/or do not have the same amplitude, are called elliptical polarization, as the projection of the electric vector traces out an ellipse as depicted in Figure 1.5c.

To describe the polarization ellipse two parameters are needed: the ellipticity angle $\chi = \arctan(a/b)$ and the orientation angle $\psi$ as indicated in Figure 1.4. The minor axis is defined as $a$; the major axis as $b$. The factor $a/b$ is referred to as the ellipticity. As such, all elliptical polarization states have an ellipticity between 0 and 1. Note that linear polarized light and circular light can be described as special cases of elliptical polarization with ellipticity 0 and 1, respectively.

### 1.2.1 Jones calculus

All the polarization information can be reduced to a single vector, called the Jones vector [7]. The Jones vector emerges from a solution of the plane wave by re-writing the electric field as:

$$E(r, t) = |E|\text{Re}\{J \exp(kt - \omega t)\}, \quad (1.3)$$

where

$$J = \begin{pmatrix} J_x \\ J_y \end{pmatrix} = \begin{pmatrix} \cos \psi \exp(i\alpha_x) \\ \sin \psi \exp(i\alpha_y) \end{pmatrix}, \quad (1.4)$$

is the Jones vector in the $(x,y)$-plane. The Jones vector can also be expressed by the orientation and ellipticity angles $\psi$ and $\chi$ as:

$$\begin{pmatrix} J_x \\ J_y \end{pmatrix} = \begin{pmatrix} \cos (\psi) \cos (\chi) - i \sin (\psi) \sin (\chi) \\ \sin (\psi) \cos (\chi) + i \cos (\psi) \sin (\chi) \end{pmatrix}. \quad (1.5)$$

This method is especially useful to describe the propagation of polarized light through optical elements such as polarizers and wave-plates. The Jones calculus also describes the optical functionality of components. A polarizer is an optical element that passes a state
Chapter 1

of polarization by absorbing or reflecting the orthogonal state. A wave-plate is an optical
element that consists of an anisotropic material, which changes the phase of transmitted light
depending on its polarization state. Common optical elements are described by a $2 \times 2$ Jones
matrix, enabling the description of the change of polarization states by multiplication of the
Jones vector of the incoming polarized light with the Jones matrix of an optical element.
The Jones vector after propagation through the optical element results. In Table 1.1 a few
examples are given of Jones vectors and Jones matrices of optical elements. In Chapter 5, the
Jones calculus is used to describe the diffraction of polarized light by anisotropic gratings.
Jones matrices will be derived for every diffraction order to describe the change of polarization
as a result of propagation through the gratings.

<table>
<thead>
<tr>
<th>Polarization</th>
<th>Jones vector</th>
<th>Optical element</th>
<th>Jones matrix</th>
</tr>
</thead>
<tbody>
<tr>
<td>Linear in $x$-direction</td>
<td>$(1 \ 0)$</td>
<td>Linear polarizer with axis of transmission horizontal</td>
<td>$(1 \ 0)\quad (0 \ 0)$</td>
</tr>
<tr>
<td>Linear in $y$-direction</td>
<td>$(0 \ 1)$</td>
<td>Linear polarizer with axis of transmission at angle $\phi$</td>
<td>$(\cos^2 \phi \quad \cos \phi \sin \phi)\quad (\sin \phi \cos \phi \quad \sin^2 \phi)$</td>
</tr>
<tr>
<td>Linear at 45°</td>
<td>$\frac{1}{\sqrt{2}} (1 \ 1)$</td>
<td>Left handed circular polarizer</td>
<td>$\frac{1}{2} \begin{pmatrix} 1 &amp; -i \ i &amp; 1 \end{pmatrix}$</td>
</tr>
<tr>
<td>Left circular</td>
<td>$\frac{1}{\sqrt{2}} (1 \ -i)$</td>
<td>Half-wave plate with fast axis in $x$-direction</td>
<td>$\begin{pmatrix} -i &amp; 0 \ 0 &amp; i \end{pmatrix}$</td>
</tr>
<tr>
<td>Right circular</td>
<td>$\frac{1}{\sqrt{2}} (1 \ i)$</td>
<td>Quarter-wave plate with fast axis in $x$-direction</td>
<td>$\exp(i\pi/4) \begin{pmatrix} 1 &amp; 0 \ 0 &amp; i \end{pmatrix}$</td>
</tr>
</tbody>
</table>

Table 1.1: A list of polarization states and optical elements with their corresponding Jones vector and matrix respectively

1.2.2 Stokes parameters

A shortcoming of the Jones Calculus is that the method does not take the intensity of light
into account. Whenever the intensity is of importance, preference is given to a method, that
describes the polarization state by means of so called Stokes parameters. Stokes parameters
are a set of values that describe the polarization state, expressed either as products of electrical
field components or as a function of orientation and ellipticity. They are defined on an
orthogonal $(x,y)$-basis in the plane of polarization as:

$$\begin{pmatrix} S_0 \\ S_1 \\ S_2 \\ S_3 \end{pmatrix} = \begin{pmatrix} E_x E_x^* + E_y E_y^* \\ E_x E_x^* - E_y E_y^* \\ E_x E_y^* + E_y E_x^* \\ i(E_x E_y^* - E_y E_x^*) \end{pmatrix} = I_0 \begin{pmatrix} 1 \\ \cos 2\psi \cos 2\chi \\ \sin 2\psi \cos 2\chi \\ \sin 2\chi \end{pmatrix}. \quad (1.6)$$

Around 1892 Henri Poincaré derived a method to facilitate the visualization of the relationship
between the Stokes Parameters and the polarization state of electromagnetic radiation,
by using the Poincaré sphere. By placing $S_1$, $S_2$ and $S_3$ on the axis of a Cartesian coordi-
nation system a so called Stokes space is created. The Poincaré sphere has its center at the
origin of this space and has a radius $S_0$. $S_0$ represents the intensity, the linear polarization
states are described by $S_1$ and $S_2$ and the circular polarization is described by $S_3$. Each point
on the Poincaré sphere represents a specific state of polarization as indicated in Figure 1.6.
Reversibly, all states of polarization can be represented on this sphere.
The polarization state and intensity of light is represented by a vector as indicated in Equation 1.6. In Table 1.2 examples of common polarization states are represented with normalized intensity ($S_0 = 1$) by their Stokes vector.

Similar to Jones calculus, optical elements can also be described by matrices, the $4 \times 4$ Mueller matrices. Multiplying a Mueller matrix by a Stokes vector, representing the intensity and polarization state of the incoming light, results in a Stokes vector describing the transmitted intensity and polarization state. Table 1.3 lists examples of Mueller matrices of optical elements.

In Chapter 5, the Stokes parameters will be used to describe the polarization-selectivity of the diffraction of anisotropic films. As will be explained, the birefringence of the diffracting material can also be described by local Stokes parameters. To make the distinction between describing the birefringence or polarization state, the lower case "s" is used for the birefringence, and the upper case "S" will be used for the polarization state.
10
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<table>
<thead>
<tr>
<th>linear polarizer</th>
<th>linear polarizer</th>
<th>linear polarizer</th>
</tr>
</thead>
<tbody>
<tr>
<td>horizontal</td>
<td>vertical</td>
<td>at +45°</td>
</tr>
<tr>
<td>$1 \begin{pmatrix} 1 &amp; 1 &amp; 0 &amp; 0 \ 1 &amp; 1 &amp; 0 &amp; 0 \ 0 &amp; 0 &amp; 0 &amp; 0 \ 0 &amp; 0 &amp; 0 &amp; 0 \end{pmatrix}$</td>
<td>$\frac{1}{2} \begin{pmatrix} 1 &amp; -1 &amp; 0 &amp; 0 \ -1 &amp; 1 &amp; 0 &amp; 0 \ 0 &amp; 0 &amp; 0 &amp; 0 \ 0 &amp; 0 &amp; 0 &amp; 0 \end{pmatrix}$</td>
<td>$\frac{1}{2} \begin{pmatrix} 1 &amp; 0 &amp; 1 &amp; 0 \ 0 &amp; 0 &amp; 0 &amp; 0 \ 1 &amp; 0 &amp; 1 &amp; 0 \ 0 &amp; 0 &amp; 0 &amp; 0 \end{pmatrix}$</td>
</tr>
<tr>
<td>half wave plate</td>
<td>quarter wave plate</td>
<td>quarter wave plate</td>
</tr>
<tr>
<td>fast-axis vertical</td>
<td>fast-axis vertical</td>
<td>fast-axis horizontal</td>
</tr>
<tr>
<td>$\begin{pmatrix} 1 &amp; 0 &amp; 0 &amp; 0 \ 0 &amp; 1 &amp; 0 &amp; 0 \ 0 &amp; 0 &amp; -1 &amp; 0 \ 0 &amp; 0 &amp; 0 &amp; -1 \end{pmatrix}$</td>
<td>$\begin{pmatrix} 1 &amp; 0 &amp; 0 &amp; 0 \ 0 &amp; 1 &amp; 0 &amp; 0 \ 0 &amp; 0 &amp; 0 &amp; -1 \ 0 &amp; 0 &amp; 1 &amp; 0 \end{pmatrix}$</td>
<td>$\begin{pmatrix} 1 &amp; 0 &amp; 0 &amp; 0 \ 0 &amp; 1 &amp; 0 &amp; 0 \ 0 &amp; 0 &amp; 1 &amp; 0 \ 0 &amp; 0 &amp; -1 &amp; 0 \end{pmatrix}$</td>
</tr>
</tbody>
</table>

Table 1.3: A list of optical elements with their corresponding Mueller matrix

1.3 Lasers

Coherent light is essential for the formation of holographic gratings, since only coherent light gives regular interference patterns. Lasers (Light Amplification by Stimulated Emission of Radiation) are commonly used as a coherent light source. For this reason, lasers will be discussed in more detail.

Lasers are oscillating amplifiers of light, emitting photons identical in phase, direction and amplitude. The working principle of a laser is based on absorption and spontaneous and stimulated emission. The conditions for a laser to work can be summed up by 1. population inversion, 2. presence of an excitation medium and 3. a resonant optical cavity, as will be elaborated in the paragraphs below.

1.3.1 Population inversion

Radiant emission and absorption take place within the atomic or molecular structure of materials. Each atom has a series of energy levels determined by the atomic orbitals. The lowest possible energy level is called the ground state. If an atom is in the ground state, it will stay there until it is excited by an external energy source. The energy level will increase by absorption and decrease by emission of a photon. The energy levels are discrete and differ by a value of $h\nu$, where $h$ is Planck’s constant and $\nu$ is the frequency of the photon.

The absorption coefficient at a given frequency is the difference between the rates of emission and absorption at that frequency. The rate of excitation from energy level $E_1$ to the next level $E_2$ is proportional to the number $N_1$ of atoms in the lower energy level and the transition probability. Similarly, the rate of stimulated emission is proportional to the population of the upper level $N_2$ and the transition probability [8]. Therefore, the absorption coefficient depends on the difference between the populations of the lower and upper level $N_1$ and $N_2$, and the flux of the incident wave.

When a material is at thermal equilibrium, nearly all atoms are in the ground state. Since the rate of absorption exceeds that of emission, the absorption coefficient at any frequency is positive. If enough light is supplied, the populations are shifted until the number of atoms in the lower and higher level are the same ($N_1 = N_2$). Under these conditions the rates of absorption and stimulated emission are equal and the absorption coefficient is zero. $N_2$ can
never exceed $N_1$ because every upward transition is matched by a downward transition. However, if more than two energy levels exist, under certain conditions an additional excitation can cause a situation where the population in the higher energy level is higher than that in the lower, $N_2 > N_1$. This condition is called population inversion.

For a four level energy transfer scheme, as indicted in Figure 1.7, a photon with frequency $\nu_1$ excites (or pumps) atoms from $E_1$ to $E_4$. If the probability of the transition of $E_4$ to $E_3$ is larger than that of the transition form $E_4$ to $E_1$ and $E_4$ is unstable, atoms will almost immediately decay from $E_4$ to $E_3$. If energy level $E_3$ is relative stable the population will grow rapidly. Eventually the atom will decay from $E_3$ to $E_2$ emitting a photon with frequency $\nu_2$. Finally, if $E_2$ is unstable, atoms will quickly fall back to ground state $E_1$. As such, the population of $E_3$ is kept large and $E_2$ low enabling a population inversion between $E_2$ and $E_3$. The absorption coefficient at frequency $\nu_2$ is therefore negative, meaning light of this frequency is amplified as it passes through the material, called an 'active medium'.

1.3.2 Excitation medium

In this thesis, most holographic experiments are carried out with an Argon ionized gas UV laser at a wavelength of 351 nm. The properties of Argon have been investigated in detail [8]. Its energy level diagram is shown in Figure 1.7. UV radiation occurs due to transitions between excited states of doubly ionized Argon ($\text{Ar}^{2+}$), which has a ground state 43 eV above that of a neutral electron. Two electron collisions or one extremely energetic single collision are required to move between the ground states. For these reasons the UV Argon laser operates at high current densities ranging from 600-1000 A/cm$^2$. The UV output rises exponentially with current density to a point beyond the design limit of the laser (3W). The output is also strongly dependent of the Argon gas pressure. For UV output the laser works best at low pressure. By reducing the number of argon atoms, the time between collision with free electrons increases and therefore the average electron energy increases.
1.3.3 Resonant optical cavity

A resonant optical cavity, which is defined by two mirrors, provides feedback to an active medium. Emitted photons parallel to the cavity axis are reflected interacting with other ions from the active medium, while stimulated emission creates two photons of equal energy, phase and direction at every interaction with the active medium. As such, light is amplified until the equilibrium between excitation and emission is reached.

The emission frequency of the laser is determined by its resonance condition. Only frequencies $\nu_m$ with a standing wave matching the optical cavity length $L$ are amplified. A discrete set of frequencies, called longitudinal-modes, is emitted which is spaced such that:

$$\Delta \nu_m = \frac{c}{2L},$$

(1.7)

with $c$ the speed of light. The coherence length, i.e. the distance over which the output beam maintains a fixed phase relationship, is inversely proportional to the spectral line-width (with $\Delta \nu$ the full width at half maximum):

$$l_c = \frac{c}{\pi \Delta \nu}.$$  

(1.8)

For example, an Argon laser (used in most of the experiments covered in this thesis), has the coherence length is in the range of meters.

1.4 Recording materials

Holographic recording is defined as the transformation of an interference pattern of lasers in photo-sensitive films. The recording in this context is the conversion of the modulations of the interference pattern to refractive index modulations in a material. Several types of recording materials exist each with there own processing, properties and applications.

Commercially available holographic materials typically consist of polymeric binders, acrylic monomers, and plasticizers, along with additives such as initiators, chain transfer agents, and photosensitizing dyes [9, 10]. In general, holographic films are benchmarked by their refractive index modulation $n_1$, defined as the effective amplitude of the refractive index modulation. These photopolymers are recorded directly by a photopolymerization reaction upon exposure to light. The high intensity parts of an interference pattern activate a photo-initiator and locally start the polymerization. The polymerization induces a diffusion of reactive and non-reactive compounds in an initially homogeneous film. Due to this diffusion differences in refractive index or differences in film thickness are created [11, 12].

Another well know class of photo-sensitive materials are the photo-resists. They have been developed for lithographic processes and record the (interference) pattern indirectly and require additional processing steps, as shown in Figure 1.8. First a photo-resist is coated on a substrate and, if needed, the solvent is evaporated in a heating step (Figure 1.8a). Next the substrate is exposed to an intensity pattern created by irradiating through a mask, writing with an e-beam or in our case with an holographic interference pattern (Figure 1.8b). Often a heating step is also needed after the exposure step to start the (de-)polymerization of the photo-resist. The advantage of this indirect recording is that this allows multiple exposures and therefore complex structures [13, 14] can be made by multiple relative simple exposures. Depending on the photo-resist, being either a positive or negative resist, the exposed part
Introduction

Figure 1.8: Photo-lithography in three steps: (a) a coating step of the photo-resist, (b) an exposure through a mask to pattern the resist and (c) a developing step to structure the resist.

is removed or remains, respectively, by a developing step (Figure 1.8c). An example of a positive photo-resist is given in Chapter 6, where it is used to etch a diffractive pattern in an Indium Tim Oxide layer.

Another indirect method to create modulations in layer thickness, also referred to as surface relief structures, is called photo-embossing [15]. In contrast to photo-resists, this method does not require a development step. Instead, the relief structure is formed with a heating step after optical exposure. Starting with a substrate coated with a monomer dissolved polymer and a photo-initiator, the substrate is exposed at a temperature below the glass transition of the polymer mixture. After the exposure, the substrate is heated above the glass transition temperature allowing the monomer to diffuse to the exposed areas to polymerize, increasing the volume and thus creating a relief structure.

Besides materials capturing the intensity pattern, also holographic recording materials exist, which capture the polarization state of light. Azobenzene polymers are the most common materials for this purpose, that induce birefringence upon irradiation.

Table 1.4 shows often used materials for holographic recording applications with their most relevant properties. The maximum diffraction efficiency depends on layer thickness, refractive index modulation, and whether or not the material absorbs light. The sensitivity of the material to light determines its exposure dose. The resolution limit, given in the table, indicates the maximal possible number of interference lines per mm of the gratings.

<table>
<thead>
<tr>
<th>Material</th>
<th>Processing</th>
<th>Max. DE</th>
<th>Exp. dose</th>
<th>Resolution</th>
</tr>
</thead>
<tbody>
<tr>
<td>Photographic emulsions [16]</td>
<td>Wet</td>
<td>50%</td>
<td>0.01 – 1 μJ/cm²</td>
<td>6000 lines/mm</td>
</tr>
<tr>
<td>Dichromated gelatin [17]</td>
<td>Wet</td>
<td>100%</td>
<td>100-600 mJ/cm²</td>
<td>1000 lines/mm</td>
</tr>
<tr>
<td>Photo-resists SU-8 [14]</td>
<td>Wet</td>
<td>100%</td>
<td>100-300 mJ/cm²</td>
<td>4000 lines/mm</td>
</tr>
<tr>
<td>Photopolymer DMP-128 [18]</td>
<td>Humid</td>
<td>80%</td>
<td>5-30 mJ/cm²</td>
<td>3000 lines/mm</td>
</tr>
<tr>
<td>Photopolymer HRF [19]</td>
<td>Dry</td>
<td>90%</td>
<td>30 mJ/cm²</td>
<td>3000 lines/mm</td>
</tr>
<tr>
<td>Azobenzene-polymer [20]</td>
<td>Dry</td>
<td>90%</td>
<td>1 mJ/cm²</td>
<td>6000 lines/mm</td>
</tr>
</tbody>
</table>

Table 1.4: Common holographic recording materials with their processing conditions, maximum diffraction efficiency, exposure dose and resolution

Unfortunately, the above described holographic materials are not very suitable for display applications. Either they are not transparent for visible wavelengths, have a low diffraction efficiency or resolution, or are not stable at room temperature.
1.5 Objectives

The performance of optical devices, such as LCDs, with respect to light efficiency, brightness and battery life is limited due to the use of absorption-based polarizers and color filters. Here, new optical elements are described based on holography, which produce polarized light with high efficiency by diffraction of one polarization direction of light and recycling the other. The diffraction of light is at near normal angles, collimated and without absorption.

Also a theoretical framework is developed to describe the polarization-selective diffraction of polarization gratings. A classification is made of the different types of polarization holograms based on the material properties and recording conditions. An initial experimental verification of the theoretical predictions is performed by measuring holographic films with a modulating birefringence.

Organic light emitting diodes (OLEDs) are inefficient due to trapping of light. Therefore, it is aimed to create gratings in the functional layers inside the OLED. The eventual goal is to improve the efficiency of the OLEDs by diffracting the trapped light.

1.6 Scope of the thesis

The performance of polarization-selective holographic films will be theoretically predicted and experimentally verified. In order to be able to make gratings with the desired optical properties, first the relevant physical and chemical aspects, such as interference, optical recording and diffraction, are described. Chapter 2, describes the theory of the formation of an interference pattern. The focus in this chapter will be on interference patterns with a modulation of intensity. The diffractive properties of recorded intensity pattern is described by several theories. These theories are compared with respect to predicting the polarization-selective diffraction.

The theoretical predictions of the diffractive properties of gratings, in particular polarization-selective diffraction, are experimentally verified in Chapter 3. Slanted holographic gratings have been made using two different approaches. In a first approach, polarization-selective diffraction is obtained for gratings with a high refractive index modulation. By selecting the proper film thickness and slant angle for each value of refractive index modulation, one direction of linear polarized light is diffracted selectively at the Bragg angle. In a second approach a refractive index modulation is obtained for only one direction of linear polarized light. Matching the refractive index of a polymer matrix with one of the refractive indices of a liquid crystal creates polarization-selective diffraction, if the LCs are aligned parallel or perpendicular to the grating direction.

Next, in Chapter 4, the rigorous coupled wave theory and Kogelnik’s coupled wave theory are compared with respect to polarization-selective diffraction. They are used to predict the influence of grating properties and to optimize grating parameters. Theoretical and experimental results are compared in order to improve the diffraction efficiency of gratings.

In Chapter 5, a different type of holography is presented, which allows the design and production of highly efficient diffraction gratings. The theory behind the unique polarization-selective diffractive properties of these gratings is derived. The polarization gratings are classified and some particular cases are modelled and experimentally verified.

In Chapter 6 gratings are investigated, which are incorporated in the functional layers of OLEDs in order improve their efficiency.
Chapter 2

Holographic Interference and Diffraction

2.1 Introduction

As discussed in the previous chapter, efficient phase gratings with a single order of diffraction of polarized light are potentially useful in LCDs as a replacement for absorbing polarizers and color filters. Here the design of such gratings is discussed on the basis of a theoretical framework, which links the diffractive properties to the grating parameters and recording conditions. As such, this chapter forms the basis for the fabrication of polarization-selective diffractive elements that are discussed in Chapter 3.

In this chapter, different types intensity and/or polarization distributions of interference pattern are examined. First, a general mathematical and graphical description of interference is given in Section 2.2. In general, two specific cases of interference patterns are distinguished from this mathematical description: purely intensity-modulated interference patterns with constant polarization state (intensity holography), and purely polarization-modulated interference patterns with constant intensity (polarization holography). This chapter addresses intensity holography. The full description of polarization holography is presented in Chapter 5.

Section 2.3 discusses the diffraction of the recorded gratings. Several diffraction theories are introduced and compared with respect to their ability to predict the diffraction efficiency of polarized light. Using these results, the diffractive properties of the different types of gratings are investigated and compared.

2.2 Interference of plane waves

Interference is the superposition of two or more waves, and can be conveniently described mathematically as the summation of plane waves. Starting with the straightforward case of two coherent plane waves, interference can be described by the Jones calculus in combination with Stokes parameters and the Poincaré sphere. Two interfering plane waves, of which the wave vectors make an angle $2\theta$, are defined by their intensities and polarization states $I_1$, $\psi_1$, $\chi_1$ and $I_2$, $\psi_2$, $\chi_2$, where $\psi_i$ is the orientation angle and $\chi_i$ the ellipticity angle of the polarization. Therefore, first the commonly used $(s,p)$-basis of the plane waves with wave
vectors $\mathbf{k}_1$ and $\mathbf{k}_2$ are defined on a global $(x,y,z)$-basis of the interference pattern:

\[
i_{s1} = i_{s2} = i_y \quad \text{(2.1)}
\]

\[
i_{p1} = -\cos \theta i_x + \sin \theta i_z \quad \text{(2.2)}
\]

\[
i_{p2} = -\cos \theta i_x - \sin \theta i_z \quad \text{(2.3)}
\]

where $i_x, i_y, i_z, i_s$ and $i_p$ are the unit vectors of the corresponding directions. The total electric field of interference $\mathbf{E}_{\text{int}}$ using Jones calculus is then defined as:

\[
\mathbf{E}_{\text{int}} = \sqrt{I_1} \exp(i \mathbf{k}_1 \cdot \mathbf{r}) \begin{pmatrix} -J_{p,1} \cos \theta \\ J_{s,1} \\ J_{p,1} \sin \theta \end{pmatrix} + \sqrt{I_2} \exp(i \mathbf{k}_2 \cdot \mathbf{r}) \begin{pmatrix} -J_{p,2} \cos \theta \\ J_{s,2} \\ -J_{p,2} \sin \theta \end{pmatrix}, \quad \text{(2.4)}
\]

where

\[
\mathbf{k}_1 = k(\sin \theta \mathbf{x} + \cos \theta \mathbf{z}), \quad \text{(2.5)}
\]

\[
\mathbf{k}_2 = k(-\sin \theta \mathbf{x} + \cos \theta \mathbf{z}), \quad \text{(2.6)}
\]

and $(J_{s,n}, J_{p,n})$ is the Jones vector describing the state of polarization of the interfering plane waves, in terms of their ellipticity angle $\chi_n$ and orientation angle $\psi_n$ (and where "s" and "p" correspond to the transverse-electric and transverse-magnetic field components in the plane wave's local coordinate system):

\[
\begin{pmatrix} J_{s,n} \\ J_{p,n} \end{pmatrix} = \begin{pmatrix} \cos \psi_n \cos \chi_n - i \sin \psi_n \sin \chi_n \\ i \cos \psi_n \sin \chi_n + \sin \psi_n \cos \chi_n \end{pmatrix}. \quad \text{(2.7)}
\]

Substituting the electric field as defined in Equation 2.4 into the definitions of the Stokes parameters (Equation 1.6) results in the following modulating Stokes parameters:
\[ S_0(x) = I_1 + I_2 + 2\sqrt{I_1 I_2} \left[ \cos(\chi_1 - \chi_2) \cos(\psi_1 - \psi_2) \cos \delta(x) - \sin(\chi_1 + \chi_2) \sin(\psi_1 - \psi_2) \sin \delta(x) \right] \]  
\[ S_1(x) = I_1 \cos 2\psi_1 \cos 2\chi_1 + I_2 \cos 2\psi_2 \cos 2\chi_2 + 2\sqrt{I_1 I_2} \left[ \cos(\chi_1 + \chi_2) \cos(\psi_1 + \psi_2) \cos \delta(x) - \sin(\chi_1 - \chi_2) \sin(\psi_1 + \psi_2) \sin \delta(x) \right] \]  
\[ S_2(x) = I_1 \sin 2\psi_1 \cos 2\chi_1 + I_2 \sin 2\psi_2 \cos 2\chi_2 + 2\sqrt{I_1 I_2} \left[ \cos(\chi_1 + \chi_2) \sin(\psi_1 + \psi_2) \cos \delta(x) + \sin(\chi_1 - \chi_2) \cos(\psi_1 + \psi_2) \sin \delta(x) \right] \]  
\[ S_3(x) = 2(I_1 \cos \chi_1 \sin \chi_1 + I_2 \cos \chi_2 \sin \chi_2) + 2\sqrt{I_1 I_2} \left[ \sin(\chi_1 + \chi_2) \cos(\psi_1 - \psi_2) \cos \delta(x) - \cos(\chi_1 - \chi_2) \sin(\psi_1 - \psi_2) \sin \delta(x) \right], \]  

with
\[ \delta(x) = 2kx \sin \theta. \]

In this case, the two wave vectors are defined to be in the \((x,z)\)-plane with angle \(\theta\) with respect to the z-axis (as indicated in Figure 2.1a). In the derivation of the modulating Stokes parameters a small angle approximation is also made (i.e. \(\sin \theta \approx 0\) and \(\cos \theta \approx 1\)). Note that for a grating with a period of 4 \(\mu\)m, recorded with UV (351 nm) the error is about 3%. Using these Stokes parameters, which are now a function of the position \(x\) along the grating on a global basis (\(i_x = i_y\) and \(i_p = i_z\)), the modulations can be visualized in Stokes space. In general, the modulation forms an ellipse, as illustrated in Figure 2.2c, whose circumference corresponds with the period of the grating. The center of the ellipse is positioned on a line through the points that represent the state of polarization of the recording plane waves as depicted in Figure 2.2c.

Two special cases can be defined representing different types of interference:

- **Intensity holography:**
  The polarization state of the interfering plane waves are equal \((\chi_1 = \chi_2 \text{ and } \psi_1 = \psi_2)\).
  In this case, both plane waves are represented by the same point in Stokes space and the ellipticity of the interference ellipse is zero. The interference pattern is therefore represented by a line. If the intensities of the plane waves are equal, this line is determined by the origin of Stokes space and twice the distance of the point representing the polarization state \((S_1, S_2, S_3)\) of the recording plane waves, as indicated in Figure 2.2a.
  In the illustrated example the polarization state of the recording plane waves has been chosen linear and horizontal and with a normalized intensity \((S_1 = 1, S_2 = 0, S_3 = 0)\).
  In the general case of intensity holography the interference is always represented by a line with the polarization state of the intensity modulation identical to that of the plane waves. Since only the intensity modulates, the recording of these interference patterns requires materials, that are sensitive to the intensity. Common recording materials form a modulation of refractive index or a surface relief.

- **Polarization holography:**
  For these interference pattern, both polarization states are 'orthogonal' \((\chi_1 = -\chi_2 \text{ and } \psi_1 = \psi_2)\).
Figure 2.2: Representations of interference patterns in Stokes space with (a) an intensity hologram, (b) a polarization hologram and (c) an arbitrary interference pattern.
ψ_1 - ψ_2 = π/2) and a line defined by the polarization states (in Stokes space) of the recording plane waves includes the origin. As shown in Chapter 5, the interference pattern in this case can always be represented by a circle on the Poincaré sphere. If the intensities of the plane waves are equal, the center of the circle is at the origin of the Stokes space, as illustrated in Figure 2.2b. In this specific example, the polarization states of the plane waves are on the S_3-axis representing right-handed and left-handed polarized light. The modulation of the interference pattern is a circle in the (S_1,S_2)-plane and, therefore, consists of only linear polarization states. As the distance of every point on the circle to the origin is constant, there is no modulation in intensity. To record this type of interference patterns materials are required, that are sensitive to the polarization state of light. Instead of forming a refractive index modulation, a modulation of birefringence is needed to record the polarization information. In Chapter 5, polarization holograms will be discussed extensively.

This chapter is focussed on intensity holography; interference patterns formed by plane waves with the same polarization state and intensity. In Section 2.2.1, the simplest case of a two-beam interference pattern is discrised. In Section 2.2.2, this is extended to a three-beam interference pattern.

Limiting ourselves to an intensity modulation implies that the polarization state of the interference pattern is always equal to that of the recording plane waves. The Jones calculus with the Stokes parameters is then not necessary. To appreciate this, consider for example the intensity modulation of an interference pattern of two linear horizontal polarized beams ($χ_1 = χ_2 = ψ_1 = ψ_2 = 0$). The modulating Stokes parameters are:

\[
\begin{align*}
S_0(x) &= I_1 + I_2 + 2\sqrt{I_1 I_2} \cos(δ(x)) \\
S_1(x) &= I_1 + I_2 + 2\sqrt{I_1 I_2} \cos(δ(x)) \\
S_2(x) &= 0 \\
S_3(x) &= 0.
\end{align*}
\]

As can be seen, both $S_0$ and $S_1$ modulate identically, which represent the intensity and the polarization state, respectively. $S_2$ and $S_3$ do not modulate and are zero. Clearly, describing the intensity modulation alone suffices. The interference modulation can be described by a summation of the electric field. The total electric field of $m$ plane waves is described by:

\[
E_{tot} = \frac{1}{2} \sum_{m=1}^{N} (E_m(r) \exp(ik_m - i\omega t) + E_m^*(r) \exp(-i k_m + i\omega t)).
\]  

The total irradiance $I(r)$, the intensity of light in a medium with dielectric permittivity $ε$ and refractive index $n$, is proportional to the squared magnitude of the total electric field integrated over time [21]:

\[
I(r) = \frac{εc}{n} \frac{1}{2} \frac{N}{\mu_0/ε_0} E(r) \cdot E^*(r) = \frac{n}{2\sqrt{μ_0/ε_0}} \sum_{l=1}^{N} \sum_{m=1}^{N} E_l \cdot E_m^* \exp(i(k_l - k_m) \cdot r),
\]
with \( c \) the speed of light, \( \varepsilon_0 \) the electric and \( \mu_0 \) the magnetic permittivities in free space. As mentioned in Chapter 1 the plane wave is described by the propagation vector \( \mathbf{k} \) and electric field \( \mathbf{E}_0 \) vectors. As they are preferentially expressed in spherical coordinates, Table 2.1 shows the conversion of the spherical to the cartesian coordinate system of the vectors.

The intensity \( A_m \) and the polarization state \( \psi_m, \delta_m \) are identical for every plane wave in the case of full intensity modulating interference. In the next sections, Equation 2.18 is applied to more specific cases; two-beam and three-beam interference patterns.

### 2.2.1 Two-beam interference patterns

The most straightforward interference pattern is constructed by two overlapping plane waves (coherent beams) with identical polarization state. Typically, this pattern is used to make holographic gratings as described in Chapter 3. It consists of a periodic sinusoidal intensity pattern as indicated in Figure 2.1. If the beams \( (\mathbf{k}_1, \mathbf{k}_2) \) are placed in the \( x,z \)-plane with an angle of \( \theta \) and \(-\theta\) from the \( z \)-axis respectively, the intensity will be independent of \( y \) and modulate in the \( x \)-direction with a periodicity determined by Bragg’s Law:

\[
\Lambda = \frac{\lambda}{2 \sin(\theta)},
\]

where \( \Lambda \) is the period, and \( \lambda \) the wavelength of the light. The modulation of the intensity \( I_A \) of the two-beam interference pattern can be derived from the Stokes parameter \( S_0 \) from Equation 2.8 and simplifies in this case to:

\[
I_A(x) = I_1 + I_2 + 2\sqrt{I_1 I_2} \cos \delta(x),
\]

where \( \delta(x) = 2kx \sin \theta \). As mentioned before, the intensity modulation is both determined by the amplitude and polarization state of the plane waves. If the polarization states are not identical, the amplitude of the intensity modulation \( I_A \) decreases with increasing angle differences \( (\psi_1 - \psi_2) \) as:

\[
I_A(x) = I_1 + I_2 + 2\sqrt{I_1 I_2} \cos(\psi_1 - \psi_2) \cos \delta(x).
\]

For angle differences of \( \pm \frac{\pi}{2} \) (orthogonal beams) the intensity modulation is zero. In this case there is still interference, but only in states of polarization as indicated in Figure 2.2b and as explained further in Chapter 5.
Holographic Interference and Diffraction

2.2.2 Three-beam interference patterns

Various interference patterns can be made using three coherent beams. The three-beam interfering patterns are for example used to pattern photo-resists for etching the ITO layer of OLEDs, as described in Chapter 6. In this thesis only 2D interference patterns, constructed with thee linear polarized plane waves equally spaced along the azimuth, are discussed. This results in a pattern with a triangular lattice. The lattice constant is only dependent on the angle $\theta$ (the angle of any of the plane waves with the z-axis) as:

$$\Lambda = \frac{\lambda}{\sqrt{3}\sin\theta}. \quad (2.22)$$

The shape of the unit cell of the interference pattern is determined by the polarization angles $\psi_m$ of the plane waves. Using equation 2.18 the intensity of a three-beam interference pattern is derived, as formulated before by Escuti et al. [21]:

$$I(x) \propto |E_1^2| + |E_2^2| + |E_3^2| + \text{Re}\{2E_1 \cdot E_2^* \exp(i(k_1 - k_2) \cdot r) + 2E_2 \cdot E_3^* \exp(i(k_2 - k_3) \cdot r) + 2E_3 \cdot E_1^* \exp(i(k_3 - k_1) \cdot r)\}. \quad (2.23)$$

If all beams are p-polarized ($\psi_m = 0$) the interference pattern consists of dark spots in an hexagonal layout as indicated in Figure 2.3a. The inverse pattern (bright spots) is generated when all beams are s-polarized ($\psi_m = \frac{\pi}{2}$) (Figure 2.3b).

2.2.3 Holographic gratings

Holography uses the interference pattern (hologram) of two or more coherent beams to record a grating in a photo-sensitive material. As elaborated further in the next chapter, one of the
main advantages of this technique over using photo-lithography (wet-etching and shadow-mask), is that different grating spacings can be obtained with a single setup. To change the grating spacing only the angle between the interfering beams needs to be changed, while mask lithography requires a new mask for every new spacing.

The diffractive properties of the holographic gratings are assumed to be constant over the entire film. In general, this is difficult to accomplish, due to the lack of homogeneity of the intensity across the interfering laser beams. In this thesis, the diffractive properties of the holographic gratings have been determined from relatively small areas, validating constant grating properties. In principle it is possible to make homogeneous, large gratings with holography with high quality beam expanders, beam splitters, mirrors and clean-room conditions.

2.3 Diffraction of isotropic gratings

So far a description of interference patterns and a way to record them in polymer materials or photo-resists has been given. In this section, the optical properties of the recorded structures are described. When light is directed through the optical structure, it diffracts. In case the optical structure is a hologram, the diffraction is called a replay, reading or reconstruction of the hologram. First, the concept of diffraction itself is explained. Second, the far-field diffracted intensities of phase and surface relief gratings are summarized, and attention is given to the differences between thick and thin gratings. Third, the two dominant theoretical formulations quantifying diffraction, the Rigorous coupled wave theory (RCWT) and Kogelnik’s coupled wave theory (KCWT), will be compared. Both theories predict the intensity and polarization state of the diffracted light in every order. Both theories are used since the RCWT is more descriptive and gives only numerical results, while the KCWT is analytical but less general. Using the KCWT, a set of grating parameters are developed that lead to polarization-selective diffraction. The RCWT will be used in Chapter 4 to model various gratings designs.

Diffraction is a phenomenon by which wavefronts of propagating waves ‘bend’ in the neighborhood of obstacles or through a slit or aperture. Two aspects are of importance: the angle and the intensity of diffraction of the transmitted orders. The angle $\theta_m$ of the $m^{th}$ diffracted order is determined by the grating equation:

$$\sin \theta_m \cos \phi_{out} = n \sin \theta_{in} \cos \phi_{in} + \frac{m\lambda}{\Lambda},$$

(2.24)

where $\theta_{in}$ and $\phi_{in}$ are the angles of the incidence of light perpendicular and parallel to the grating structure (lines) respectively (as indicated in Figure 2.4) and $m = 0, 1, 2...$ the diffraction order. The diffraction intensity cannot be analytically calculated in a general way, as it depends on the grating and material parameters. However, the diffraction efficiency of the transmitted orders $\eta_m$ can be defined in a straightforward way as:

$$\eta_m = \frac{|D_m|^2}{\sum_{m=0}^{\infty} |D_m|^2},$$

(2.25)

with $D_m$ the electric field of the $m^{th}$ forward propagating diffraction order.

In this chapter, a scalar diffraction theory will be used, where the intensity of light is represented by a value. The polarization state of light is neglected. However, in Chapter 5, the polarization state will be included in the diffraction theory by using a vector notation.
instead of a scalar value. To derive diffraction efficiencies of periodic structures, the diffraction
of a single aperture or slit is derived first. A grating is then modeled as a periodic and infinite
set of slits. The intensity of diffraction around apertures can be described approximately by
a mathematical formalism called scalar diffraction theory.

Following the derivation described by [22, 23], one starts by solving the Helmholtz equation. Depending on the distance $R$ of the aperture from the point of observation, either the
Fraunhofer or Fresnel regime applies. The distinction between both regimes is made by the
Fresnel number ($F$).

\[ F = \frac{\Lambda^2}{\lambda R}. \] (2.26)

In this thesis only Fraunhofer diffraction ($F \ll 1$) is described, as it applies to the optical
far-field properties of gratings (arrays of wavelength size apertures). In this regime, the
diffraction pattern is independent of the distance $R$ to the screen and a small angle approxi-
mation is made (paraxial diffraction). The interference pattern scales with the distance of the
screen to the aperture since it depends only on angles. Let the coordinates in the aperture
plane be $(x', y')$ and the coordinates in the projection plane $(x, y)$, as illustrated in Figure 2.5.
Writing the aperture function as: $\xi(x', y')$, the Fresnel-Kirchhoff diffraction integral gives the
electric field at the projection plane as:

\[ U(x, y) = A \int_{aperture} \xi(x', y') \exp(-ik(xx' + yy')/R)dx'dy', \] (2.27)

where $k$ is the wavenumber, $R$ is the distance from aperture to the projection plane, $A$ is a
scalar value, and the integral is taken over the aperture. A diffraction grating is a set of parallel
slits. Let there be $N$ equal slits with a transmittance function of $\xi(x')$, which is repeated
periodically with period $\Lambda$. To obtain the resulting diffraction pattern the integration needs
to be extended over all $N$ periods. It may therefore be written as:

\[ U(u) = \sum_{n=0}^{N-1} A \int_{-\infty}^{\infty} \xi(x') \exp(-iku(x' + n\Lambda)) dx' \]

\[ = A \left[ \sum_{n=0}^{N-1} \exp(-ikn\Lambda u) \right] \int_{-\infty}^{\infty} \xi(x') \exp(-iku x') dx', \] (2.28)
2.3.1 Diffraction of phase and amplitude gratings

The general transfer function of a grating is described by:
where \( n(x) \) is a complex function, which describes the refractive index and absorption profile of a single period of the grating. An approximation is made that the refractive index is does not change in the \( z \)-direction and is constant over the layer thickness \( d \). The real part of the function \( n(x) \) describes the optical density or refractive index. The imaginary part describes the absorption of the grating. If the function is purely real, the grating is called a phase grating. If it is purely imaginary, it is called an amplitude grating. Amplitude gratings are known to be less efficient in terms of diffraction. As such, phase gratings are more commonly used. For only very few gratings, \( T(x) \) has an analytical solution, with block shaped gratings and sinusoidal gratings being the most common. The analytical solutions of the diffraction efficiencies of these gratings are listed in Table 2.2. The diffraction of phase gratings without an analytical solution can be solved numerically (i.e. with Fast Fourier Transform (FFT)).

<table>
<thead>
<tr>
<th>Grating Profile</th>
<th>Transfer function</th>
<th>Diffraction Efficiency</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sinusoidal</td>
<td>( \exp\left(-i2\pi n_1 \cos(2\pi x/\Lambda)\right) )</td>
<td>( J_m^2(\frac{2\pi n_1}{\lambda}) )</td>
</tr>
<tr>
<td>Binary</td>
<td>( (\exp(i2\pi n_1) + \exp(-i2\pi \Lambda u))\text{sinc}(\frac{\Lambda u}{2}) )</td>
<td>( 4\cos^2(\frac{\pi \Lambda m}{2}) + 2n_0^2\text{sinc}^2(\Lambda m) )</td>
</tr>
</tbody>
</table>

Table 2.2: Some grating profiles with their corresponding Fourier transform and diffraction efficiency

### 2.3.2 Diffraction of thin and thick gratings

So far, the diffraction efficiency \( \eta \) has been considered independent of the polarization state of the incoming light. Light has been treated as a scalar; a single value representing the intensity. For thin gratings this approximation is usually valid. Therefore, thin phase or amplitude gratings are not suitable to diffract polarization selectively.

Thick gratings change the state of polarization during the propagation of light through the grating. Several diffraction theories exist to describe this change of polarization state and in the coming Sections 2.3.3 and 2.3.4 two of them will be explained in more detail.

The distinctive Klein parameter \( Q \) between thick and thin gratings depends besides on the layer thickness \( d \), on the wavelength of the irradiance \( \lambda \), the average refractive index \( n_0 \) and the periodicity of the grating \( \Lambda \), as [24]:

\[
Q = \frac{2\pi \Lambda d}{n_0 \Lambda^2}. \tag{2.35}
\]

If \( Q < 1 \) the grating is considered to be thin, whereas \( Q > 10 \) the grating is considered to be thick. Otherwise, the theories of thin and and thick gratings do not apply. The distinction can physically be determined by the number of diffraction orders. Thick gratings have only one order of diffraction (Bragg diffraction), while thin gratings have many (Raman-Nath diffraction). Polarization gratings are a notable exception to this, since they can have only one diffraction order while still being thin-gratings in the classical sense (see Chapter 5).

Another important difference between thick and thin gratings is the angle dependency of the diffraction efficiency \( \eta \). While the diffraction efficiencies \( \eta_m \) for thin gratings are relatively insensitive to incident angle, thick gratings diffract only in a small angular interval around the Bragg-angle \( \theta_B \):
\[
\sin \theta_B = \frac{\lambda}{2n_0 \Lambda}. \quad (2.36)
\]

### 2.3.3 Kogelnik’s Coupled Wave Theory

One of the most popular descriptions of thick gratings is the Kogelnik’s Coupled Wave Theory [25] (KCWT). It is well suited to describe polarization-selective diffraction for slanted thick gratings. In this section the main aspects of KCWT are elaborated.

The theory is built around the condition that only two orders propagate inside the grating: the first order (+1) and the zeroth order (0). This means that the theory is valid for thick gratings. With this condition, the electric field inside the hologram results from the superposition of the electric fields of the two orders. By solving Helmholtz’ equation, two differential equations are obtained that are related by a coupling constant. An analytical approximation of the differential equations yields the diffraction and transmission efficiencies for phase and amplitude gratings. For mixed gratings, with the absorption modulation and the refractive index modulation are in phase, the diffraction efficiency can be considered as consisting of separate contributions of both gratings.

Additionally, this theory assumes that the refractive index \( n \) and/or absorption coefficient \( \alpha \) modulation is sinusoidal along the grating as described by:

\[
n = n_o + n_1 \cos(g \cdot r), \quad (2.37)
\]

\[
\alpha = \alpha_0 + \alpha_1 \cos(g \cdot r). \quad (2.38)
\]

This assumption of a sinusoidal grating is not quite as limiting as it seems, since many complicated profile can be modeled by considering the superposition of multiple sinusoidal gratings [25].

For a thick grating, Kogelnik’s general formula 2.39 for the diffraction amplitude \( D_A \) of a transmission grating determines the diffraction efficiency \( \eta \) at the Bragg-angle and Bragg-wavelength. For non-slanted gratings \( (\varphi_G = 0) \) the \( \eta \) is independent of polarization direction; for slanted gratings the \( \eta \) is determined for s and p polarized light separately.

\[
D_A = -i \sqrt{\frac{c_R}{c_S}} \exp(\xi) \frac{\nu^2 \sin(\nu^2 - \xi^2)}{\nu^2 - \xi^2}, \quad (2.39)
\]

\( \nu \) is the modulation parameter, which is dependent on the polarization direction, \( \xi \) is called the dephasing parameter, which expresses the deviations from the Bragg condition \( (\Delta \theta, \Delta \lambda) \) and \( c_R \) and \( c_S \) are the obliquity factors:

\[
\nu_s = \frac{2\pi n_1 d}{\lambda \sqrt{|c_R c_S|}}, \quad \nu_p = \frac{2\pi n_1 d}{\lambda \sqrt{|c_R c_S|}} \cos(2\theta_B - 2\phi_K), \quad (2.40)
\]

\[
\xi = \frac{1}{2} d \left( \frac{\alpha}{c_R} - \frac{\alpha}{c_S} - i \frac{\nu}{c_S} \right), \quad (2.41)
\]

\[
c_R = \cos \theta_B, \quad c_S = \cos \theta_B - \frac{\Lambda}{\lambda} \cos \varphi_G. \quad (2.42)
\]

The degree of dephasing is represented by \( v \).
Figure 2.6: Diffraction efficiency at Bragg-conditions of s- and p-polarized light as a function of $dn_1$ of a slanted grating with slant-angle of $23^\circ$, period $\lambda$ of 450 nm and average refractive index 1.55. The grey line indicates the polarization contrast (p/s).

$$v = \frac{2\Delta \theta \pi}{\Lambda} \sin(\varphi_G - \theta_B) - \frac{\Delta \lambda \pi}{n_0 \Lambda^2}.$$  \hspace{1cm} (2.43)

Finally, the diffraction efficiencies of both linear polarization directions $\eta_{s,p}$ of the grating is determined by:

$$\eta_{s,p} = \sqrt{c^2} D_A D_A^*.$$  \hspace{1cm} (2.44)

Polarization-selective diffraction from a slanted grating is achieved by choosing a combination of layer thickness $d$ and refractive index modulation $n_1$ for a slanted grating. As $d$ or $n_1$ continues to increase, the diffraction efficiency reaches a maximum as follows from Figure 2.6 and starts to decrease again. The dependency of the diffraction efficiency as a function of $dn_1$ is different for s- and p-polarized light (also indicated in Figure 2.6). The decrease of intensity for increasing $n_1$ is called "over-modulation".

Since the diffraction efficiency $\eta$ of both s- and p-polarized light as a function of $d$ and $n_1$ are different, a combination is chosen in such a way that one of the polarization direction has a low $\eta$ and the other a high $\eta$. In Figure 2.6 as an example the $\eta$ (at Bragg-conditions) of both s- and p-polarized light at 611 nm are shown for a slanted grating with a period $\Lambda$ of 450 nm and slant angle $\varphi_G$ of $23^\circ$ as a function of $dn_1$. The polarization-contrast (p/s) has been indicated by the grey line in Figure 2.6. As can be seen for $dn_1$ values of 0.48, i.e. a grating with layer thickness of 20 $\mu$m and $n_1$ of 0.024 $\mu$m or layer thickness of 10 $\mu$m and $n_1$ of 0.048 only p-polarized light is diffracted. Note, s-polarized light is diffracted selectively for $dn_1$ values of 0.82 $\mu$m.

Although the KCWT is easy to use and gives a good first analytical approximation of the $\eta$ for Bragg-gratings, it has some limitations. First of all, the modulation of refractive index is always considered to be sinusoidal. This is equivalent to a first order approximation of any periodic refractive index modulation. Typically, not all periodic refractive index modulations
are sinusoidal. Second, the theory is only valid for linear polarized light parallel and perpen-
dicular to the grating. Third, the evanescent waves are neglected, and fourth, the theory is
only valid for small energy exchanges between coupled waves, i.e. for low absorptions.

2.3.4 Rigorous Coupled Wave Theory

Another theoretical framework to calculate the diffraction of thick gratings is the Rigorous
Coupled Wave Theory (RCWT). The RCWT is a straightforward technique to obtain the
exact solution of Maxwells equations for the electromagnetic diffraction by grating structures.

Gaylord, Moharam et al. [26] developed a stable and efficient numerical implementation
of this theory. The implementation is a non-iterative, deterministic technique using a state-
variable method that converges to the proper solution without inherent numerical instabilities.
The accuracy of the solution obtained depends mainly on the number of terms in the field
space-harmonic expansion as explained below. In this theory the grating is divided into 3
regions, above, below and at the grating, as indicated in Figure 2.7:

- Region I: On one side (i.e. the top) of the grating, the incoming electrical field is
indicated by \( E_{\text{in}} \), the reflection of the diffracted orders are indicated by \( R_i \). Usually,
the refractive index of air is chosen in this region. The total electric and magnetic fields
are given by:

\[
E_I = E_{\text{in}} + \sum_i R_i \exp(-i(k_{xi}x - k_{I,zi}z)),
\]

\[
H_I = \left( \frac{i}{\omega \mu} \right) \nabla \times E_I,
\]

where \( k_{xi} \) is the \( x \)-component of wave vector the diffracted orders \( i \) and \( k_{I,zi} \) the \( z \)-
component of the wave vector of the reflected orders.

- Region II: On the other side of the grating, only transmitted orders \( E_{II} \) are present,
having a uniform refractive index, without correspondence to the refractive index of air
nor the grating.

\[
E_{II} = \sum_i T_i \exp(-i(k_{xi}x - k_{II,zi}(z - d))),
\]

\[
H_{II} = \left( \frac{i}{\omega \mu} \right) \nabla \times E_{II},
\]

with \( k_{II,zi} \) the \( z \)-component of wave vector of the transmitted orders \( i \).

- Region G: The grating region has a constant thickness, is bound by Region I and II,
and has a periodic refractive index modulation. The tangential electric (\( y \)-component)
and magnetic (\( x \)-component) fields are expressed using a Fourier expansion of the
space-harmonic fields:

\[
E_G = \sum_i S_{yi}(z) \exp(-ik_{xi}x),
\]
\[ H_G = -i \sqrt{\frac{\varepsilon_0}{\mu_0}} \sum_i U_{xi}(z) \exp(-ik_{zi}x). \] (2.50)

The general approach for solving the exact electromagnetic boundary value problem associated with the diffraction grating is found by solving the Maxwells equations in each of the three regions, followed by matching the tangential electric- and magnetic-field components at the boundaries of the three regions.

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{division_of_single_layer_binary_grating.png}
\caption{Division of a single layer binary grating in regions.}
\end{figure}

Without further details of the implementation, for which it is referred to at [26], the electromagnetic field is determined with the help of the Fourier transform of the refractive index modulation of the grating. For every diffraction order the tangential electric (y-component) and magnetic (x-component) field is solved at the boundaries of the grating using a 2 × 2 matrix with eigenvalues and eigenvectors. The diffraction efficiencies \( \eta_i \) of the transmitted orders are calculated, when all the transmission and reflection orders are known, using:

\[ \eta_i = T_i T_i^* \Re \left( \frac{k_{II} z_i}{n_{II}} \left/ \left( \frac{k_0 \cos(\theta)}{n_I} \right) \right. \right), \]

(2.51)

where \( T_i \) are the transmitted orders, \( n_{II} \) the refractive index of the grating material.

By extending the theory from a 1D layer to a layered stack, it can also be applied for surface relief gratings, slanted gratings or any periodic type of grating. A layered stack is constructed as indicated in Figure 2.8. The theory for these multi-layered gratings has been implemented and shown to be stable by [27].

The solutions to the multi-layer grating diffraction are found by the following sequence. First, the coupled-wave equations are constructed and are solved for the electromagnetic fields in each grating layer identical to a single layer. Second, the electromagnetic boundary conditions (continuity of the tangential electric and magnetic-field components) are applied to the input region I and the first grating layer \( d_1 \). Next, the boundary conditions are applied to all consecutive pairs of grating layers, followed by applying them to the last grating layer \( d_L \) and the output region II. Finally, the resulting array of boundary condition equations is solved for the reflected and the transmitted diffracted field amplitudes. Thus the diffraction efficiencies are obtained.
Every layer of the stack needs to be described by a periodic function in $x$ of the refractive index. In Chapter 4, this implementation is used to model the diffraction of several gratings. The fast Fourier transform (FFT) is used to determine the Fourier harmonics. The total 2D refractive index modulation of a stack is represented as a matrix. Increasing number of matrix elements improves the accuracy.

![Multi-layer stack of gratings](image)

**Figure 2.8:** Multi-layer stack of gratings

The disadvantage of the RCWT is that it only yields numerical answers and no analytical formulas, which give more insight. In contrast to the KCWT, which is only valid for thick gratings, the RCWT is also applicable to thin gratings and gratings with $1 < Q < 10$. An additional advantage of RCWT is that it is able to describe the diffraction of any periodic modulation, not only sinusoidal modulations. Furthermore, the RCWT can also describe the diffraction of not only linear polarization light but also circular and elliptical polarized light.

### 2.4 Discussion and Conclusions

Interference, in its most general way, has been described as a modulation of Stokes parameters. A distinction has been made between polarization interference and intensity interference. For intensity interference, it has been shown that a description of the modulation in intensity is sufficient. The interference patterns of two and three plane waves have been described in terms of angle, wavelength and polarization state.

Next, the concept of diffraction was introduced. The diffraction of thin gratings under the paraxial approximation has been partially derived and it has been shown that these gratings are not polarization-selective. A similar vector-based diffraction theory will be used as the basis to describe the diffraction of polarization gratings in Chapter 5. For the diffraction of thick gratings, two theories are introduced to predict the polarization-selective diffraction: the rigorous coupled wave theory (RCWT) and Kogelnik’s coupled wave theory (KCWT).

The Rigorous Coupled Wave Theory (RCWT) is a straightforward numerical technique for obtaining the exact solution of Maxwell’s equations for an electromagnetic wave propagation through a grating. Two main advantages of this theory over the KCWT can be identified.
First, the RCWT applies to all isotropic transmission gratings, regardless of their classification into thin or thick regimes (i.e. for any Q). Second, contrary to KCWT, the RCWT can be used to effectively describe non-sinusoidal profiles, especially those profiles that have variation in the transverse dimension of the grating (relative to the grating vector). The advantage of the KCWT is that it gives analytical solutions for the diffraction efficiency. This gives more insight in the diffraction dependencies of the grating parameters. The RCWT gives only numerical answers.

In Chapter 3, the RCWT is used to fit the measured diffraction efficiency as a function of angle in order to estimate the refractive index modulation of thick and thin gratings and gratings in between both regimes. The RCWT is also extensively used in Chapter 4 to determine the influence of the various grating parameters, such as the layer thickness and the refractive index profile. The KCWT is used to determine the grating parameters to obtain polarization-selective diffraction of thick slanted gratings.
Chapter 3

Polarization-selective out-coupling from waveguides

3.1 Introduction

In the previous chapters theoretical aspects of holography and diffractive gratings have been explained. It has been shown [5], how under certain conditions a slanted grating can diffract polarization-selectively by using a specific combination of layer thickness and refractive index modulation amplitude.

In the next sections, two types of gratings are experimentally verified to diffract with polarization-selectivity, strong collimation and with minimal absorption. First, the diffractive properties of thick isotropic gratings will be shown. Requirements to couple out (diffract) polarization-selective waveguided light are met, as predicted by Kogelnik’s coupled wave theory (KCWT). As previously mentioned by Jagt et al. [5], by combining a modest refractive index modulation ($n_1$) of 0.03 with a matching layer thickness $d$ of about 20 $\mu$m, $p$-polarized light can be coupled out, while $s$-polarized light remains waveguided. Secondly, polarization-selective diffraction of anisotropic gratings will be shown in Section 3.3. By aligning liquid crystal molecules along the grating direction in a polymer matrix, polarization-selective diffraction occurs on the basis of matching the $n_o$ and mismatching the $n_e$ of aligned liquid crystals with the isotropic refractive index of the polymer.

Slanted gratings have been recorded in specially developed photopolymers with a holographic intensity pattern using a UV-transmission setup. In Section 3.2.3 and 3.3.2 the polarization-selective out-coupling has been measured using the glass substrate as a waveguide. The high refractive index modulation gratings and the anisotropic gratings are compared and discussed in Section 3.4

3.2 Out-coupling using high refractive index modulation

As mentioned before, holographic materials require recording of holographic optical elements at UV or visible wavelengths and typically consist of polymeric binders, acrylic monomers, and plasticizers, along with initiators, chain transfer agents, and photo-sensitizing dyes [9, 10]. In general, holographic films are benchmarked by their refractive index modulation (RIM) $n_1$, defined as the effective amplitude of the first term of the Fourier expansion of the refractive index profile of the grating period. Usually, only the first term of the Fourier expansion is
taken into account (sinusoidal refractive index modulation is assumed), in accordance with Equation 2.37. The amplitude of the RIM of thick gratings can be experimentally determined by fitting a coupled wave theory (Kogelnik or RCWT) to measured diffraction efficiencies ($\eta$) of transmission gratings.

The HRF materials from DuPont [28] ($n_1 \approx 0.07$) and the DMP 128 of Polaroid [29] ($n_1 \approx 0.08$) are two high refractive index grating materials that are widely studied [30, 31, 32] and commonly used. The high $n_1$ values of Dupont materials are based on the refractive index differences between a polymeric matrix and the monomer diffusing components. The high $n_1$ of Polaroid materials originates from the formation of air voids formed through a complex wet processing after holographic exposure. In display applications, losses of light in the visible range due to absorption should be avoided and usually holograms with a small pitch (< 1$\mu$m) and a high $n_1$ are required. Unfortunately, both previously mentioned materials are less suitable in displays because of their lack of transparency. It is therefore desirable to generate holographic materials with a high $n_1$ that can be recorded with UV-light, while preserving transparency in the visible range.

In the following, the formation of a highly transparent porous holographic grating with a high refractive index modulation ($n_1 \approx 0.14$) and modest thickness ($\approx 20\mu$m) will be demonstrated. This high refractive index modulation is obtained using a simple three-step process and a composition comprising of a UV-sensitized mixture consisting of acrylates, an epoxy monomer and an inert, non-polymerizable, compound, each of the components having a specific function.

3.2.1 Processing and recording

The objective is to create a grating consisting of a periodic structure of alternating porous and solid polymer. Thereto, a process is developed consisting of the following steps. First, a holographic UV-exposure records a phase grating in a cell, filled with a homogenous mixture, by polymerizing the acrylates in the bright areas of the interference pattern. This initiates diffusion of acrylate monomers to the bright areas, where it is consumed by the polymerization process. Also a counter diffusion occurs of the less reactive epoxy monomer and the inert compound to the dark area. Secondly, a flood UV-exposure polymerizes the multifunctional epoxy to form a second cross-linked polymer network that phase separates from the inert compound into nano-porous elements. Finally, the cell is opened, enabling the inert compound to evaporate and leaving a transparent modulated film with alternating predominantly an acrylate network with a nano-porous epoxy network. Because the nano-pores reduce the refractive index of the epoxy-rich areas, the refractive index modulation is relatively large.

Our base material (mixture) is composed of four primary components mixed at equal weight percent (25 wt% each): a pentacrylate monomer, a diacrylate monomer, an inert compound and an epoxy monomer.

Multifunctional acrylates (25wt% di-pentaerythritol penta-acrylate (DPPA) and 25wt% poly(ethylene glycol)(200)di-acrylate (PEGDA) are used because of their high reactivity and fast gelation in combination with a small amount (1%) of free-radical photoinitiator (Irgacure 184). The DPPA reacts within seconds and forms a dense cross-linked network. The (PEGDA) copolymerizes with DPPA, acts as a softener, and helps to dissolve the different components and to form a homogeneous mixture prior to polymerization.
Toluene (25 wt%) is used as the inert volatile solvent, which evaporates within seconds after opening of the cell. This is observed by recording the diffraction efficiency during the evaporation process, i.e. initially a low diffraction is observed and the diffraction efficiency increases with increasing evaporation of the volatile solvent. During its evaporation, toluene is replaced by air, thus forming a porous structure. The volume percentage of the pores is determined by the amount of inert compound (in our case the solvent).

**The epoxy monomer**, a diglycidylether of bisphenol A, is chosen because of its low shrinkage. In the presence of a cationic photo-initiator it cures under UV-exposure into a densely cross-linked network, preventing the collapse of the porous structure. The porous structure is further supported by the acrylate walls at each side. The cross-link density of both the acrylate and epoxy network together with the polymerization kinetics influence the pore sizes. These pores remain small enough (see Figure 3.1) to avoid scattering of visible light, therefore the film remains transparent. We have estimated scattering losses in our holographic gratings by comparing the intensity of the incident beam with the sum of the diffracted (zero and +1 orders) and reflected beams (about 3% of scattering was observed).

To avoid rupture of the film on opening of the cell, one of the glass slides of the cell
is coated with a mono-layer of ((3-glycidoxypropyl)trimethoxysilane) [33]. During the flood exposure the epoxy-polymer reacts with the glycidyl groups of this covalently bonded silane improving the adhesion of the film to the substrate. The epoxy-polymer adheres so strongly that it leaves residual lines once the holographic film is removed from the glass substrate. This is clearly seen in the SEM images of Figure 3.1a and c. It also demonstrates that the counter diffusion of the epoxy monomer took place during the holographic exposure, concentrating the epoxy in the dark areas. In supplementing experiments the opposite glass substrate was coated with a mono-layer of trichloro(tridecafluoro-1,1,2,2-tetrahydrooctyl) silane to increase the difference in adhesive strength between both sides of the cell even further.

After filling a cell with the photo-reactive mixture a hologram is recorded with a 351 nm UV-laser (60 s, 50 mW/cm² each beam). The evolution of the first order of diffraction of a typical recording is shown in Figure 3.2. The initial delay in the diffraction growth is attributed to oxygen inhibition of the free-radical polymerization by oxygen dissolved in the monomer mixture. Once the oxygen is consumed by its reaction with the free-radicals generated by the photo-initiator, polymerization starts in an effective way building up the holographic grating. Two recordings are shown with a periodicity Λ of 500 nm. The differences in diffraction efficiency (intensity) is caused by a difference in layer thickness between both samples.

Next, the cell is flood exposed with a 365 nm UV lamp for one hour, ensuring maximum conversion of the epoxy and acrylate monomers. The flood exposure time is indicated in Figure 3.2 by the two vertical striped lines. Finally, the cell is opened enabling the toluene to evaporate and produce a holographic film with a high refractive index modulation on a single glass substrate. The moment of opening is also indicated in Figure 3.2 by the dotted vertical line on the right-side.

At the beginning of the evaporation the diffraction efficiency increases for both recordings. However, for the thick \( d = 20 \mu m \) sample the intensity of the first order of diffraction starts
3.2.2 Analysis of the refractive index modulation

To determine the RIM, the diffraction efficiencies $\eta_0$ and $\eta_1$ of $s$- and $p$-polarized light of wavelength 633 nm as a function of angle $\Psi$ have been measured for non-slanted gratings with a period of 520 nm, 2080 nm and 4510 nm and a layer thickness of 12.4 $\mu$m, 4.0 $\mu$m and 3.6 $\mu$m respectively, as illustrated in Figure 3.3b. The refractive index is assumed to have a sinusoidal modulation along the grating vector with amplitude $n_1$. The grating with periodicity of 520 nm meets the conditions of a thick or Bragg grating ($Q = 122$). The grating with periodicity of 4510 nm meets the conditions of a thin or Raman-Nath grating ($Q = 0.50$) and the grating with periodicity of 2080 nm is neither thin nor thick ($Q = 2.6$). The $n_1$ of these three gratings have been determined differently.

To determine the amplitude of the RIM $n_1$ of the thick grating, the first order of diffraction $\eta_1$ of $s$-polarized light of 633 nm as a function of angle $\Psi$ is measured and fitted using the RCWT. The best fit, using a least square error method, for a non-slanted transmission grating with a layer thickness of 12.4 $\mu$m, a period of 520 nm and average refractive index of 1.497 was achieved for a sinusoidal RIM with an amplitude of 0.047 as shown in Figure 3.3a.

For gratings in between the thick and thin regime, the refractive index modulation has been determined by fitting the diffraction efficiencies of the diffracted orders using a least square error method as seen in Figure 3.4. The average refractive index $n_0$ and the amplitude of the RIM $n_1$ of a grating with thickness of 4.0 $\mu$m and periodicity of 2.08 $\mu$m have a best fit for values of 1.470 and 0.076.

Similarly, the amplitude of the RIM $n_1$ of a thin grating with a periodicity of 4510 nm be determined by fitting the measured diffraction efficiencies to the theoretical values. The best fit is obtained results in an average refractive index of 1.39 and modulation amplitude of 0.064 using the least square error method. As can be seen the fit is less accurate as the previous case as the higher diffraction orders do not appear in the fit. In Chapter 4 it will be shown that by using a different (non-sinusoidal) grating profile a better fit can be achieved.

Figure 3.3: a) Measured and fitted intensity of diffraction from a 633 nm laser as a function of angle for a 12.4 $\mu$m thick non-slanted grating with a 520 nm period and a refractive index amplitude of 0.047 and b) the measurement setup.

to decrease after about 1 minute of opening the cell due to over-modulation, as explained in Section 2.3.3.
Alternatively, the refractive index amplitude can be estimated by the measured ratio \( \eta_1/\eta_0 \). This ratio is 0.315 and corresponds to a theoretical \( n_1 \) of about 0.10. This has been obtained using Equation 3.1 for thin sinusoidal gratings as previously listed in Table 2.2:

\[
\eta_i = J_i^2\left(\frac{\pi n_1}{\lambda}\right).
\]

3.2.3 Out-coupling from nano-porous slanted gratings

As mentioned in the introduction of this chapter, one potential application of these high RIM and porous grating is the selective out-coupling of polarized light from a waveguide, which can be used for backlight systems in displays. Kogelnik’s coupled wave theory (KCWT) predicts the diffracting efficiency \( \eta \) of \( s \)- and \( p \)-polarized light from a slanted Bragg grating. Similar to the work of Jagt [5], slanted gratings were recorded in transmission by UV-light in such a way, that visible light is coupled out at near normal angles making use of the wavelength dispersion of the grating.
Figure 3.5: Holographic recording geometry and resulting grating.

Slanted gratings were recorded using the transmission geometry with the 351 nm line of an Argon ion laser (50 mW/cm² each beam). The recording angles (+71.5° and +13.4° degrees, as shown in Figure 3.5) were adjusted in order to have Bragg diffraction of green light at the normal.

In this way a slanted transmission grating with period Λ of 450 nm and slant angle $\varphi_G = 23°$ was recorded in films of thickness $d = 21$ μm. The SEM pictures in Figure 3.6 shows a cross-section of a recorded slanted grating with a 450 nm period and slant angle close to 23° (actually a larger slant angle is obtained due to a small collapse of the layer). In Figure 3.6b the porous structure in the grating is clearly visible showing the dense acrylate regions and porous epoxy regions.

The polarization-selectivity of this grating in the waveguiding mode was measured using this 21 μm thick film on top of a glass substrate that acts as a waveguide. Unpolarized light from a cold cathode fluorescent lamp (CCFL) is coupled into the waveguide at the edge (see Figure 3.7b). The opposite side of the CCFL of the waveguide has been painted black to avoid reflections. By measuring the light intensity as a function of angle for both polarizations of light, coupled out at near normal angles (see Figure 3.7a), polarization contrasts ($p/s$) of 7.5, 35 and 11.2 were measured for 611 nm, 546 nm and 436 nm respectively. These specific wavelengths have been chosen as they are dominantly emitted by the phosphors of the CCFL. The angular color dispersion is a result of the wavelength dependency of the Bragg angle. It is shown that red, green, and blue wavelengths are coupled out at 13°, 2°, and −13° respectively.

3.3 Out-coupling based on aligned liquid crystals

In this section, we describe anisotropic holographic slanted gratings based on polymer dispersed liquid crystals (PDLCs), that achieve a highly selective polarization behavior in a different fashion [34, 35]. The molecular shape of liquid crystals (i.e. rod- or disk-like) result in multiple refractive indices. The different refractive indices, or anisotropy, arise only from alignment of liquid crystals on a macroscopic scale. The orientation of the LCs can be induced by a flow, an orientation layer, or an electric field. The basic operation of the PDLC slanted grating is illustrated in Figure 3.8, where unpolarized light coming from a CCFL is coupled into the waveguide from the side of the cell. While total internal reflection (TIR)
Figure 3.6: SEM images of a cross-section of a slanted nano-porous film

Figure 3.7: Out-coupling of waveguided light from a slanted grating: a) intensity of $p$-(upper curve) and $s$-(lower curve) polarized light as a function of angle for both polarization direction of 611 nm (red), 546 nm (green) and 436 nm (blue). b) measurement setup.
occurs at one face of the waveguide, the hologram on the opposite face diffracts highly collimated \(p\)-polarized light (matching the Bragg condition in angle and wavelength) out of the waveguide in the normal direction, and leaves the other polarization state unaffected. This \(s\)-polarization state can be recycled by subsequently converting it to \(p\)-polarization either by a uniaxial birefringent film laminated to the waveguide or by birefringence within the waveguide itself.

In general, PDLC gratings are composed of Liquid Crystal (LC) domains within a rigid polymer binder [36]. However, in some PDLC mixtures [37], a special alignment condition can occur, where the nematic LC molecules align parallel to the grating vector to create a highly anisotropic refractive index modulation (illustrated in Figure 3.8). If the refractive index of the polymer matrix is matched to the ordinary index of the LC, diffraction of \(s\)-polarized light is very low \((n_1s \approx 0)\). Strong diffraction occurs for \(p\)-polarized light due to the high refractive index difference between the polymer matrix and the extra-ordinary index of the LC \((n_{1p} \geq 0.05)\) has been observed [38]).

The use of matching and mismatching of refractive indices has two key advantages: first, the polarizing operation of the hologram does not require a high refractive index modulation (since \(s\)-polarization is suppressed by a low \(n_{1s}\) regardless of the magnitude of \(n_{1p}\)); second, the polarization contrast \((\eta_{1p}/\eta_{1s})\) is not as sensitive to variations in the hologram thickness \(d\) compared to thick nano-porous gratings.

3.3.1 Processing and recording

An optical element, shown in Figure 3.8, comprises of two parallel glass substrates confining the holographic film. A mixture of PN393 pre-polymer [35], TL205 nematic high \(\Delta n\) LC mixture \((n_o, n_e) = (1.527, 1.745)\) at 589 nm, 20 °C) from Merck, and an additional cross-linker trimethylolpropane trimethacrylate from Aldrich were prepared with wt% ratio of 40/50/10, respectively. A refractive index of 1.485 was found for polymerized PN393 with the additional cross-linker at a wavelength of 589 nm, using an Abbe refractometer at room temperature.

A cell was constructed with 7 \(\mu m\) fiber spacers, sealed with adhesive on two sides, and filled, by capillary action, with the mixture. Slanted gratings were recorded using transmission geometry with the 351 nm line of an Argon ion laser (25 mW/cm\(^2\) each beam). The recording angles (+71.5 and +13.4 degrees, similar as shown in Figure 3.7b) were chosen in order to
have Bragg diffraction of green light perpendicular to the cell. As such a slanted transmission grating with period $\Lambda$ of 450 nm and slant angle $\varphi_G$ of $23^\circ$ was recorded in films with a thickness of 7$\mu$m. In this experiment, the glass cell used during recording of the PDLC grating itself, was used as waveguide.

After the holographic exposure, the sample was flood exposed (11 mW/cm$^2$, 30 min, 300-450 nm) to polymerize any remaining monomer. The edge opposite the CCFL was painted black to minimize reflections.

### 3.3.2 Out-coupling from slanted gratings

The highly anisotropic diffraction of this PDLC grating can be seen in Figure 3.9, where the transmission spectrum (0th order) was measured for orthogonal linear polarizations with normal incidence ($\Psi = 0^\circ$). At the Bragg wavelength of 578 nm, $p$-polarized light was diffracted into the waveguide with an efficiency of $\sim$30% while $s$-polarized light was diffracted at $\sim$2.7% efficiency (after compensating for incoherent scattering and the air-glass losses). Based on these efficiencies, a polarization contrast ($\eta_{lp}/\eta_{ls}$) of $\sim$11 can be expected when the hologram is illuminated from the edge by the CCFL. From Equation 2.44 $\eta_{lp} = 0.0184$ and $\eta_{ls} = 0.0036$ are estimated, revealing a further optimization of exposure conditions and/or materials to reduce $\eta_{ls}$ is worthwhile.

The optical performance of the backlight assembly was measured using the setup illustrated in Figure 3.7b. Light coming from a CCFL was coupled into the waveguide from the side of the cell. The polarized intensity of the emitted light was measured in the plane containing the grating vector and substrate normal with a DMS 703 (Autronic-MELCHERS GmbH, Germany) equipped with a diode array spectrophotometer. The detection angle $\Psi$ is measured from the normal direction. In order to analyze the polarization contrast of the out-coupled light, measurements for both polarizations were done with a conventional polarizer sheet on the top of the sample.

The relative intensity of red, green, and blue light (611, 546 and 436 nm, respectively) emitted by the CCFL from the hologram/waveguide assembly is shown as a polar plot in Figure 3.11, to illustrate the collimation and angular dispersion at once. In this figure, only

**Figure 3.9:** Transmittance of the slanted anisotropic hologram in PDLC materials for $p$- and $s$-polarizations.
Figure 3.10: Measured relative intensities of both orthogonal polarizations for red (611 nm), green (546 nm), and blue (436 nm) wavelengths. Also shown is the angular distribution of forward and backward emission for red. Black and gray lines correspond to s- and p-polarizations and forward and backward emissions of unpolarized light, respectively.

p-polarized light is shown and the three primary colors correspond to the three strongest emission peaks of the CCFL. Further detail is provided in Figure 3.10, where the relative intensity for both s- and p-polarizations is shown as a function of observation angle for the three colors. It can be seen that p-polarized light is emitted with much higher efficiency compared to s-polarized light for all three wavelengths. Polarization contrasts ($\eta_p/\eta_s$) of 12, 11, and 8 were measured for red, green and blue respectively. However, if the non-zero background, attributed to incoherent scattering, is discarded, the polarization contrasts are as high as 19, 18, and 16.

Even though imperfections in alignment of the liquid crystals induce a small amount of scattering, the propagation lengths in the waveguide are still large (on the order of 10s of centimeters). Further note that the angular distribution of emission is narrow due to the Bragg grating. It has been observed for all three wavelengths that the emission intensity drops to 50% of the maximum within approximately $\pm 3^\circ$.

In addition to the polarized and collimated emission, we also observe highly unidirectional emission (the difference in intensity emitted towards the viewer compared to emitted away from the viewer) from the backlight due to the $23^\circ$ slant of the holographic grating. In order to measure this, the side of the waveguide opposite the CCFL was made absorbing with flat black paint (measured reflectivity < 0.3%). Shown in the bottom right of Figure 3.10 is the intensity of 611 nm light emitted from the front and back sides of the backlight. At its peak angle, the intensity ratio ($I_{\text{front}}/I_{\text{back}}$) was 10; by subtracting the non-zero background caused by incoherent scattering, this ratio was as high as $\sim 25$. Similar results were observed for the 436 and 546 nm peaks. It is assumed, that the light emitted in the backward direction is primarily due to reflection of the forward travelling diffracted order at the glass-air interface.
Figure 3.11: Measured angular distribution of $p$-polarized emission for red (611 nm), green (546 nm), and blue (436 nm) wavelengths, highlighting collimation and angular dispersion.

on the front surface, and could be improved by adding an appropriate anti-reflection coating.

3.4 Discussion and conclusions

Two types of holographic UV-recording materials were obtained, that diffract polarization-selective from a waveguide by a slanted grating: a nano-porous material, that has a high refractive index modulation, and a polymer dispersed liquid crystals material, that has a modulation in concentration of aligned LCs. Both materials meet the requirements for LCD backlighting e.g. highly polarized, collimated, transparent and unidirectional emission. This integrates several functions typically embodied in separate optical films in a conventional LCD design. Potentially new backlight designs can consist of fewer layers and have higher emission efficiency.

Experimental results of forming a hologram recorded with 351 nm holography have been obtained. Our primary findings for the nano-porous gratings with $d = 21 \, \mu m$ with $n_1 = 0.05$ include polarization contrasts ($p/s$) of 7.5, 35 and 11 for 611 nm, 546 nm and 436 nm respectively. The large wavelength dependency of the polarization contrast for the nano-porous gratings is undesirable for display applications.

The wavelength dependency of the polarization contrast is evident by determining the over-modulation of a grating for different wavelengths. In Figure 3.12 the diffraction efficiency is shown as a function of $dn_1$ for $s$-polarized light for wavelengths 436, 546 and 611 nm from the model grating. To obtain high polarization contrasts, $\eta_s$ should be zero. The $dn_1$ values for the three wavelengths range from 0.35 to 0.5 (for the first over-modulation). To obtain polarization-selective diffraction for each wavelength, the thickness $d$ or the amplitude of the RIM $n_1$ should vary for every color in such a way that the condition of $\eta_s = 0$ is met for each color.

For display applications, a (practical) solution would be a holographic film with a different film thickness for every color pixel. To obtain high diffraction efficiency for the three colors, thicknesses of 21.5 $\mu m$, 24.9 $\mu m$ and 26 $\mu m$ for 436 nm and 536 nm and 611 nm respectively are required (for a grating with $\Lambda$ of 450 nm and slant angle of $\varphi$ of 24.2°). Since the diffraction is wavelength dependent, color separation takes places. The slanted grating is designed in such a way the green color of 536 nm diffracts at the normal of the waveguide, the red color
Polarization-selective out-coupling from waveguides

Figure 3.12: The diffraction efficiency as a function of $dn_1$ for s-polarized light of wavelengths 436, 546 and 611 nm from a grating with period $\Lambda$ of 450 nm, slant angle $\varphi$ of 24.2° and thickness $d$ of 20 $\mu$m and average refractive index $n_0$ of 1.55.

Figure 3.13: A slanted grating on a waveguide with a lens array and color filters on top. The lens array redirects the red (R), blue (B) and green (G) at a normal angle to the corresponding color pixel.

of 611 nm diffracts at a positive angle; the blue color at a negative angle. In order to have a collimated emission of all three wavelengths at the normal angle, a micro-lens array is required, as indicated in Figure 3.13. To avoid loss of color saturation by small mismatches in alignment between the diffracted rays and the color pixels, color filters may be used. Note that in this way, in addition to their improved polarization efficiency, the efficiency of the display is improved substantially, because the loss in the color filters are minimized.

It is important to note that the slanted grating couples out waveguided visible light, while the recording of this slanted hologram was performed at UV-wavelength. This facilitates the recording procedure and avoids the use of coupling optics (which is important in view of an industrial process) necessary to achieve the more oblique recording angles when longer recording wavelengths are used. Only visible wavelengths propagating within the TIR angles of the waveguide meet the Bragg condition to diffract in the direction nearly perpendicular to the substrate. This recording technique limits the slant angle to be not much higher than 23°. To couple out the green color at the normal of the waveguide the period needs to be
around 450 nm. This period determines the angular spread of other colors. As shown for the nano-porous gratings the thickness of the layer in combination with the amplitude of the refractive index modulation determines the polarization contrast. Thereby, all the parameters of the grating are fixed.

For the PDLC gratings polarization contrasts of $\sim 10$ were measured for all three wavelengths. Contrary to the nano-porous gratings, the contrast is not sensitive to wavelength. This is caused by the fact that the linear birefringence of the liquid crystals is almost constant for the whole range of visible wavelengths. Therefore, the thickness of the PDLC gratings can be arbitrarily chosen. As will be further investigated in Chapter 4, the thickness gives control over the collimation of the diffraction.

The optical quality of both gratings suffers from small amounts of scattering. For the nano-porous gratings, the pore size remains small enough to almost avoid diffuse scattering of visible light (about 3% of scattering) and the polymer film appears fully transparent. Higher concentrations of the inert compound in the initial composition lead to larger pore sizes and result in more scattering. Toluene has been chosen as inert compound for its volatility, but non-volatile non-reactive compounds in combination with an extraction process (e.g.: supercritical CO$_2$ extraction) can be used as well. Further optimization of the concentrations of all the compounds might not only avoid scattering problems but also increase the refractive index modulation even further. The PDLC gratings scatter light due to liquid crystal droplet formation. The droplets scatter at the domain boundaries and the formation of the droplets should be avoided by optimizing the concentration of all the compounds or by changing the recording conditions such as intensity and temperature.

The described PDLC gratings contain 50% LC, making it gel-like. This is undesirable with respect to their mechanical and thermal properties. For the PDLC gratings it should be possible to attain a fully polymerized film by using a reactive LC, for example using LC molecules with epoxide groups \cite{39}. The liquid crystals should exhibit the same kind of diffusion and reorientation during the holographic exposure, but could be subsequently polymerized through a thermal- or photo-initiation process with kinetic properties sufficiently different from the acrylate curing kinetics.

Examining the nano-porous gratings, the choice of a sinusoidal refractive index profile might not lead to an accurate determination of the RIM, as explained in Section 3.2.2. The gratings are created by acrylates, an epoxy, and pores, all with significant different refractive indices. A sinusoidal modulation is assumed which seems not to result in a good fit between the measured and calculated diffraction. Using another non-sinusoidal refractive index profile might lead to a better fit of the measured diffraction, as will be shown in Chapter 4.
Chapter 4

Modeling of gratings

4.1 Introduction

The refractive index profile of gratings is modeled in this chapter using the rigorous coupled wave theory (RCWT) and Kogelnik’s coupled wave theory (KCWT). Since the KCWT is limited to sinusoidal profiles and thick gratings, only the RCWT is used to explore non-thick gratings with various profiles. The relevant parameters of various profiles are determined to fit the angular diffraction efficiencies to the measurements from Chapter 3. Contrary to the previous chapters, where the amplitude of refractive index modulation was determined, and the focus was on polarization-selective diffraction, in this chapter the influence of the refractive index profile on the diffraction efficiency is determined. Instead of the diffraction efficiencies of gratings with sinusoidal refractive index modulations, the diffraction efficiencies of triangular and block-shaped gratings are investigated. The goal of this comparison is to come to a better fit of the experimentally observed diffraction efficiencies to the refractive index modulation of the grating.

In Chapter 3 polarization-selective diffraction has been demonstrated for both slanted nano-porous and PDLC gratings. The polarization-selectivity at the Bragg angle of nano-porous gratings was in approximate agreement with the predictions of the KCWT, as the exact refractive index modulation is not known. The angular and polarization-selective diffraction predictions of the KCWT will be compared to the predictions of the RCWT. Furthermore in Section 4.2.2, the RCWT will be used to predict the diffraction properties of slanted PDLC gratings with different layer thicknesses and birefringences.

As mentioned in Section 2.3.3, the KCWT is only valid for Bragg gratings. As such, the RCWT was used to fit the measured non-slanted non-Bragg gratings of Chapter 3. In that chapter a sinusoidal RIM profiles was used to fit the diffraction measurements. In this chapter, other profiles will be explored for non-Bragg gratings in order to improve upon the sinusoidal fit. Slanted nano-porous and PDLC gratings are modeled in Section 4.2 with similar grating parameters as described in Chapter 3. In Section 4.3 non-slanted gratings are modeled. The thick and non-thick gratings are discussed separately, as they behave differently with respect to profile changes. Besides the sinusoidal profile, a triangular, and several square profiles are investigated. As will be discussed in Section 4.3.2, one of the square profiles gives a better fit of the refractive index profile for nano-porous gratings to the diffraction measurements from
4.2 Modeling nano-porous and PDLC slanted gratings

Throughout this thesis, the focus has been on the design and implementation of slanted diffraction gratings for out-coupling collimated, polarization-selective, visible light from a planar waveguide. The motivation for this has been to improve the optical efficiency of LCD systems, as described in Chapter 1, and involves holographic fabrication, optical design, and materials-development. Two slanted gratings, introduced in the previous chapter, are here studied and modeled from the theoretical point of view in order to identify optimal design parameters. First, the nano-porous slanted gratings are modeled in Section 4.2.1 with the RCWT and the KCWT. Second, the PDLC gratings are modeled in Section 4.2.2 for various layer thicknesses with the RCWT.

4.2.1 Nano-porous gratings modeled with KCWT and RCWT

The requirements for designing a nano-porous polarization-selective diffractive grating allow for much freedom to choose the grating parameters. The grating of H.J.B. Jagt et al. [5] has proven to be suitable (at least for isotropic materials) and is therefore chosen. This reference grating, with a period of 450 nm, slant angle of 24°, thickness of 20 μm, average refractive index 1.55 and a sinusoidal RIM amplitude \( n_1 \) of 0.03, couples out visible light polarization selective at near normal angles.

To model a slanted angle with the RCWT, the layer is divided in sub-layers. Each sub-layer has a 1D-sinusoidal profile and is shifted with respect to the next sub-layer in the direction of the grating, as schematically shown in Figure 4.1a. The slanted grating is modeled by subdividing the layer into typically 200 sinusoidal sub-layers. The shift of each sub-layer is chosen in such a way, that an angle of 24° is constructed. The diffraction efficiency is calculated for a range of transmitted angles \( \theta \) for specific wavelengths and a slanted grating with a sinusoidal refractive index profile.

The angular dependence of s- and p-polarized light, coupled out from the reference slanted grating, has been calculated using the RCWT and KCWT, as shown in Figure 4.2a and b, respectively. The polarization contrasts for three wavelengths have been calculated. Wavelengths 436 nm (blue), 546 nm (green) and 611 nm (red) are chosen as they are the common
wavelengths within a CCFL in most LCDs (and are consistent with the measurements in Chapter 3). As can be seen from Figure 4.2, both theories predict a polarization dependent diffraction for these wavelengths. The polarization contrasts are listed in Table 4.1. Although the exact amplitude of the refractive index modulation of the measured slanted grating from Section 3.2.3 is not exactly known, both theories do not correspond to these experimentally obtained results. There is a wavelength dependency observed experimentally, but the theoretical predictions show a stronger influence of wavelength and angle for the polarization contrast.

Table 4.1: A list of polarization contrasts at the Bragg angle obtained with KCWT and RCWT for blue (436 nm), green (546 nm) and red (611 nm).

<table>
<thead>
<tr>
<th>Wavelength</th>
<th>KCWT $I_p/I_s$</th>
<th>RCWT $I_p/I_s$</th>
</tr>
</thead>
<tbody>
<tr>
<td>436 nm</td>
<td>1.4</td>
<td>0.21</td>
</tr>
<tr>
<td>546 nm</td>
<td>14</td>
<td>1.9</td>
</tr>
<tr>
<td>611 nm</td>
<td>0.9</td>
<td>4.2</td>
</tr>
</tbody>
</table>

The KCWT predicts a sinc-like decay for increasing distance from the Bragg angle, while the RCWT shows a more irregular and asymmetric finite-thickness fringes. The irregularity is a numerical artifact, caused by the discontinuous refractive index profile of the subdivisions and the asymmetry of the slanted grating. The measurements of the nano-porous gratings (shown in Figure 3.7) do not show these finite-thickness fringes, because the grating contains...
small imperfections across its transverse dimension and therefore not all periods are identical. Both theories are limited to gratings with identical grating periods.

Although the results from the KCWT and RCWT are very different form the experimentally determined diffraction efficiencies, the theories can still be useful to predict polarization-selective diffraction based on over-modulation (high $dn_1$ values). Both theories give an indication for the grating parameters such as slant angle, period and average refractive index on the angular intensity and wavelength distribution on the polarization-selective diffraction.

### 4.2.2 Collimated diffraction of slanted PDLC gratings

PDLC gratings have a separate RIM for $s$- and $p$-polarized light, based on the matching of the refractive index of aligned LCs with a polymer matrix, as described in Section 3.3. Compared to the nano-porous gratings they have a polarization contrast over a broader range of wavelengths and the polarization contrast is not sensitive to layer thickness. Under perfect refractive index matching and alignment conditions, the RIM amplitude is zero and no diffraction occurs for $s$-polarized light. For the other polarization direction ($p$-polarized light) the RIM is not zero and any value of $d\Delta n$ results in a selective-diffraction of $p$-polarized light for a broad range of wavelengths. Note that the $\Delta n$ is the effective refractive index modulation resulting from the birefringence of the LCs, and does not necessarily has the magnitude of the molecular birefringence.

As the PDLC gratings use the matching and mismatching of the refractive index of LC molecules to diffract polarization-selective, the thickness is free to choose. This freedom to choose the layer thickness allows to control the angular spread of the diffraction or collimation. A thicker grating will give a narrower angular distribution; the diffraction will be more collimated. Using the RCWT for gratings with a thickness of 1.0 $\mu$m, 7.0 $\mu$m and 16 $\mu$m the out-coupling has been numerically determined at a wavelength of 611 nm, as illustrated in Figure 4.3.
As can be seen in Figure 4.3a, the width of the diffraction peak decreases with increasing layer thickness. A width at half maximum diffraction efficiency of 7.9°, 2.7° and 0.8° is found for a thickness of 1.0 μm, 7.0 μm and 16 μm respectively. The maximum diffraction efficiency remains more or less constant for the three layer thicknesses in the case of equal \(d\Delta n\). The value of 0.1288 has been chosen similar to the experimentally determined value of the HPDLC gratings in Section 3.3. To obtain a broad wavelength interval with high diffraction efficiency an thin layer is needed requiring high \(\Delta n\). If \(\Delta n\) is taken as a fixed material parameter (like the experimentally obtained value of 0.0184), the layer thickness influences both the width of the diffraction peak and the diffraction efficiency, as seen in Figure 4.3b. The diffraction efficiency of the grating with a thickness of 1.0 μm has a diffraction efficiency of less than one percent. For out-coupling applications with broad angular diffraction distributions these gratings should have therefore a higher molecular birefringence or a better alignment.

4.3 Modeling refractive index profiles of non-slanted gratings

So far, only sinusoidal refractive index modulating gratings have been modeled. For a first order approximation of realistic gratings this is appropriate. However, in photopolymer recordings, where diffusion of multiple components takes place with different refractive indices, this refractive index profile is not likely. Therefore, more complex models of gratings deserve further exploration. In this section, the influence of the diffraction efficiency will be explored for several refractive index profiles of Bragg and non-Bragg gratings (thick gratings with \(Q > 10\), thin gratings with \(Q < 1\) and gratings with \(1 < Q < 10\)). For the average refractive index and refractive index modulation values are used that are close to the measured values of the nano-porous gratings of Chapter 3.

The refractive index of a grating is described by a periodic function. Next to the sinusoidal shaped profiles, the triangular and square shaped profiles are likely candidates to explore further. These shapes can be used as building blocks to describe more complex gratings, that are a better approximation of actual profiles of diffractive films.

4.3.1 Refractive index profiles

The non-slanted nano-porous Bragg gratings, measured in Chapter 3, were so far fitted with a sinusoidal profile to obtain the RIM amplitude. To investigate the influence of the shape of the RIM, the diffraction efficiency of the first order of diffraction has been calculated for a sinusoidal, triangular and square-shaped grating (with \(n_0 = 1.497\), \(n_1 = 0.047\), \(\Lambda = 0.520 \mu m\) and \(d = 12.4 \mu m\) at 633 nm s-polarized light). As indicated in Figure 4.4, the sinusoidal and square-shaped Bragg gratings have a thickness of 1.0 μm has a diffraction efficiency of less than one percent. For out-coupling applications with broad angular diffraction distributions these gratings should have therefore a higher molecular birefringence or a better alignment.

For gratings with \(1 < Q < 10\) the influence of the refractive index profile was also determined. As can be seen in Figure 4.5, the diffraction efficiency is different for the three basic shapes. Comparing the diffraction efficiencies of the first order of diffraction (Figure 4.5a), the triangular shaped refractive index profile has the highest diffraction efficiency, followed by the sinusoidal shaped grating. Although the angular dependencies are very different, all of the gratings have there maximum diffraction efficiency at 9°, as determined by Bragg’s law: \(\sin(\theta) = \lambda/(2\Lambda)\).
Chapter 4

Figure 4.4: (a) The diffraction efficiency of the first order $\eta_1$ of s-polarized light of a sinusoidal, square and triangular shaped grating (as indicated in b) with average refractive index $n_0 = 1.497$, refractive index amplitude of modulation $n_1 = 0.047$, period $\Lambda = 0.520 \ \mu m$ and layer thickness $d = 12.4 \ \mu m$ of 633 nm light.

If we look at the zeroth order of diffraction (Figure 4.5b), the angular response of the efficiency for the triangular-, sinusoidal- and square-shaped gratings are different. Characteristically, they have a minimum at different angles. The triangular shaped grating has its minimum at $10^\circ$, the sinusoidal shaped grating at $14^\circ$ and the square-shaped grating at $19^\circ$. This position is determined by the cumulative diffraction efficiencies of the non-zero orders (multiple diffraction orders exist since it is not a thick grating).

Comparing the modeled angular diffraction efficiencies to the measured results, as shown in Figure 4.6a, indicates that a square-shaped profile has the closest fit of the three basic shapes. Therefore, in the next section a closer look will be given to the influence of various grating parameters of a grating with a square-shape profile on the diffraction efficiency.

The angular diffraction efficiency of thin gratings ($Q < 1$) has not been calculated or measured since the diffraction efficiency is almost independent (a constant) as a function of angle. At large angles measuring errors occur due to scattering and reflections from the grating or substrate. A better way to determine the refractive index profile of thin gratings is by fitting the transmitted diffraction orders at normal incidence, as will be shown in the
Figure 4.6: a) The measured angular diffraction intensities $I_1$ and $I_0$ of $s$-polarized light of 633 nm light of a nano-porous grating with $\Lambda = 2.08 \mu m$ and $d = 4.0 \mu m$ together with the modeled results of a similar block-shaped grating using the RCWT. b) The RIM profiles of a square profile approximated with a Fourier series with $i = 1, 3, 5$ and 7.

next section.

4.3.2 Square-shaped gratings

Holographic phase gratings form their refractive index profile due to diffusion of reactive and non-reactive compounds. Therefore, the refractive index profile is expected to modulate continuously along the grating. A square-shaped (discontinuous) refractive index profile is therefore not very likely. To model a continuous square-shaped profile, it can be approximated by its Fourier series. Starting from a sinusoidal profile, the refractive index is stepwise transformed into a square-shape by means of adding higher order Fourier terms, as indicated in Figure 4.6b. The Fourier series of a square-shape profile is:

$$f(x) = n_0 + n_1 \sum_{i=1,3,5,...}^{\infty} \frac{1}{i} \sin \left( \frac{2\pi ix}{\Lambda} \right)$$

(4.1)

The diffraction efficiencies ($\eta_0$ and $\eta_1$) as a function of angle $\theta$ has been calculated for gratings with a period $\Lambda$ of 2.08 $\mu m$, layer thickness $d$ of 4 $\mu m$ and RIM amplitude $n_1 = 0.2$ with of square shapes with $i = 1, 3, 5$ and 7. Figure 4.7 shows the zeroth and first order of diffraction of s-polarized light ($\eta_{0s}$ and $\eta_{1s}$) as a function of angle $\theta$ for these profiles. As expected, the more Fourier terms are taken into account, the better the square-shape is represented and the more the diffraction efficiency spectra will be alike. Adding only one extra Fourier term ($i = 3$), results in a nearly identical diffraction efficiency spectrum as the binary square-shaped grating. Adding higher Fourier terms are therefore not required. A realistic polymer phase grating with an approximately square-shaped refractive index profile can therefore be accurately modeled by a binary (discontinuous) square profile.

A holographic phase grating commonly consist of several diffusion and non-diffusion compounds with different refractive indices. These compounds are not necessarily present in equal
volume fractions. Assuming that the refractive index profile has a nearly square-shaped refractive index profile, the so-called fill factor \( f \) does not have to be 50%. Therefore, different values of \( f \) of a square-shaped grating have been modeled, as indicated in Figure 4.8b. Again the same grating parameters are used as previously described. The diffraction efficiency of the zeroth order is shown in Figure 4.8a for several fill factors \( f \). Increasing \( f \) from 37.4% to 62.6% results in two main trends, indicated by arrows. First, the diffraction efficiency at the normal decreases as the fill factor increases. Second, there is a shift of the minimum from 13° to 21°. Especially the position of this minimum can be used to estimate the (effective) fill factor of a grating. If we look at the non-porous and measured grating in Figure 4.6, a minimum at 17° is observed. This corresponds to a fill factor \( f \) of 44.3%.

Previously in Section 3.2.2 the refractive index modulation of thin gratings (with periodicity of 4.51 \( \mu \text{m} \)) was determined by fitting the diffraction efficiencies of the diffracted orders. The assumed sinusoidal refractive index modulation resulted in a poor fit for the diffraction efficiencies of the diffracted orders. To improve this fit a block-shaped grating profile is as-
Figure 4.9: The refractive index (dotted line) and its modulation amplitude (grey marking) as a function of the fill factor of a block-shaped grating. The least square error (solid black line) has a minimum at 0.74.

sumed and the fill factor is varied. Again the least square error method is used to determine the fill factor that gives the best fit for an average refractive index and its modulation. In Figure 4.9 the fitted refractive index and its modulation are plotted together with the error as a function of the fill factor. As can be seen, a best fit (the lowest error) is obtained for a fill factor of 0.74 with an average refractive index of 1.46 and a modulation of 0.12. Note that in this case the refractive index modulates between 1.52 and 1.28 as indicated in Figure 4.10a. Contrary to the sinusoidal fit, the block-shaped refractive index profile gives also a good agreement with the higher orders of diffraction, as indicated in Figure 4.10b.

4.4 Conclusions and discussions

The diffraction efficiency of slanted Bragg gratings have been modeled by both KCWT and RCWT. The s-polarization direction of the RCWT shows a much more noisy behavior of the diffraction efficiency as a function of angle than that of the KCWT. This is most likely due to the subdivision of the layer and that the asymmetry of the slanted grating is taken into account. The subdivisions create a refractive index which is discontinuous in the $z$-direction, leading to reflection and diffraction of the propagation diffraction orders. Increasing the amount of sublayers to 2000 does not lead to a decrease of the noisy behavior. Probably, the cumulative numerical errors are in the same order of magnitude. Comparing the polarization-selectivity at the Bragg angle and angular dependency of the diffraction efficiency of sinusoidal gratings for both theories, a wavelength dependence is observed, just like the measured nanoporous slanted gratings. The main advantage of the RCWT is, that it is not limited to thick gratings ($Q > 10$) or sinusoidal refractive index profiles.

The relevance of the polarization contrasts obtained with both theories is limited, since they are only valid for the Bragg condition. Small deviations of this condition in angle or wavelength change the contrasts significantly. These angular contrast changes are caused by the finite-thickness fringes. Fortunately, the measurements do not show these finite-thickness fringes and the polarization contrast is much more constant, presumably due to impurities and small fluctuations in grating properties from period to period. The developed theories do not allow to model impurities in gratings or small fluctuations in grating properties per period. Therefore, the RCWT and KCWT are useful to estimate the gratings parameters for
polarization-selective diffraction at Bragg angles.

Also PDLC gratings have been modeled with the RCWT. Assuming perfect refractive index matching for one polarization direction of light only the diffraction of the other polarization needs to be calculated. Due to the polarization-selectivity for a broad range of wavelengths, the thickness can be chosen freely. The width of diffraction has been modeled for various thicknesses. Relative thin gratings give broad diffraction peaks, but require a high birefringence for an efficient diffraction. Thick gratings give highly collimated light and give already a high diffraction efficiency for low birefringences. For some display applications, high collimation is desirable when colors need to be separated, as discussed in Chapter 3. This can be realized in the thick gratings, which diffract highly collimated and angular wavelength dependent. Other display designs make use of white diffracting light. In this case color separation is undesired and relative thin gratings should be used. If a broad diffraction peak of 436 nm overlaps with a broad peak of 611 nm, white light is produced. For example, in Figure 4.11 the modeled angular diffraction of a thick slanted PDLC grating is shown for red (611 nm), green (546 nm) and blue (436 nm) colors with a thickness $d$ of 1.76 μm, period $\Lambda$ of 0.60 μm, $\Delta n$ of 0.13 and with a slant angle of 18.8°. As can be seen the angular diffraction of the different colors overlap each other, the color separation is small and polarized white light is produced.

The RCWT has been used to investigate the influence of the refractive index profile on the diffraction efficiency. For thick Bragg gratings the diffraction efficiencies seems to be independent of the shape of the index profile. The triangular shaped grating gave a lower diffraction efficiency, but this can be contributed to the effective amplitude of the refractive index modulation. To model the diffraction efficiency of thick gratings, the assumption of a sinusoidal shape is sufficient, and therefore the KCWT is more appropriate and easy to use.

Several methods have been used to determine the refractive index profile for different gratings. For thick gratings ($Q > 10$) the shape refractive index profile seems to be of minor
influence compared to the amplitude of refractive index modulation. The best way to fit the amplitude is by fitting the angular distribution of the first order of diffraction as shown in Chapter 3. In order to determine the refractive index profile of gratings with $1 < Q < 10$, their diffraction efficiency of the zeroth order should be measured. For thin gratings ($Q < 1$) the best way to fit the refractive index profile is by using the many diffraction efficiencies of the transmitted diffracted orders at normal incidence.

For gratings with $1 < Q < 10$ three basic shapes (triangular, square and sinusoidal) have been used to fit, as they give characteristic diffraction efficiencies as a function of angle. As an example, the refractive index profile of a non-Bragg nano-porous grating with $n_0 = 1.42$, $n_1 = 0.20$, $\Lambda = 2.08 \, \mu m$ and $d = 4.0 \, \mu m$ has been fitted to a square-shape. This shape is used, as it resembles the measured angular response most. A discontinuous (binary) square shaped grating can be used to model accurately as it shows similar results to continuous square shaped profiles. The position of the minimum diffraction efficiency of the zeroth order (of s-polarized light) has been fitted using a fill factor of a square-shaped grating and a fill factor of 44.3% was found.

Fitting the fill factor gives insight in the refractive index profile of nano-porous grating. A higher amplitude of the effective refractive index modulation can be obtained by a fill factor of 50%. Therefore, by increasing the volume fraction of the high refractive index compounds by 5.7% an increase of the the effective refractive index modulation $n_1$ is expected.

For thin nano-porous grating with a pitch of $4.51 \, \mu m$ a better fit has been obtained by using a block shaped the refractive index profile with a fill factor instead of a sinusoidal profile. The best fit was obtained for a fill factor of 0.74, average refractive index of 1.46 and refractive index modulation of 0.12. A higher effective refractive index modulation is expected, when more low refractive index components are used in the holographic recording material.

The difference of optimum fill factor for the two nano-porous gratings originates from the diffusion processes during recording. The diffusion speed and distance is balanced by the hindrance due to increase in viscosity during polymerization and a driving force for diffusion induced by concentration differences. The diffusion of monomers can be in the direction

![Figure 4.11: Modeled angular diffraction for red (611 nm), green (546 nm) and blue (436 nm) colors of a PDLC grating with a thickness $d$ of 1.76 $\mu m$, period $\Lambda$ of 0.60 $\mu m$, $\Delta n$ of 0.13 and with a slant angle of 18.8° using the RCWT.](image)
of high intensity regions of the interference pattern, but also towards the dark region, as published by C.M. Leeuwis [12]. A different grating period has therefore a different optimum composition for a maximum refractive index modulation.
Chapter 5

Polarization holography

5.1 Introduction

Polarization holography has many similarities to intensity holography. Both are pattern-transfer techniques that make use of the interference of coherent (laser) light. The period of the holographic structure is determined by the angle and the wavelength of the overlapping plane waves. To record the interference pattern, a light sensitive material is placed into the interference pattern and a corresponding pattern is somehow imprinted (through various mechanisms) in the material. Depending on the material, several processing steps are required, including development, post-curing and desensitizing steps. This yields a periodic refractive index pattern, which diffracts light.

As explained in Chapter 2 the difference between polarization holography and intensity holography is the type of modulation. Intensity holography is the modulation of intensity generated by two plane waves with the same polarization. For polarization holography two plane waves interfere with orthogonal polarization states resulting in a modulation of the polarization state, not the intensity. In this chapter it will be shown that polarization holograms diffract polarization-selectively. It will be shown how an interference pattern of polarization states can be recorded to form a modulation of linear and circular birefringence and how this results in the polarization-selective diffraction of an arbitrary state of polarization. In the previous chapters the described gratings diffracted only linear polarization-selectively.

Although several researchers have introduced theoretical studies on the diffraction of anisotropic gratings before [40, 41, 42, 43, 44], in this chapter a more complete description will be made. Starting with the formation of the general interference pattern of two arbitrary but orthogonal polarization plane waves, the formation of both linear and circular birefringent gratings is described. Afterwards, the polarization-selective diffraction properties in terms of Stokes parameters of the recorded polarization gratings are derived. With this theoretical framework the polarization-selective diffraction can be predicted using the polarization states of the recording beams and the material sensitivity to polarized light.

In Section 5.2 a general description in terms of Stokes parameters is given for interference patterns of polarization gratings. Furthermore, the interference of specific orthogonal polarization states are described: linear horizontal and linear vertical polarized, linear orthogonal at $\pm 45^\circ$ and right- and left-handed circular. It will be shown that any polarization hologram can be represented by a circle on the Poincaré sphere.

In Section 5.3 a theoretical framework is derived to describe the diffractive properties
of polarization gratings. The paraxial diffraction theory is used, as described in Chapter 2, with a vectorial diffraction notation. The complex dielectric permittivity tensor \([42, 41]\) of the material is used as the transfer matrix in terms of modulating Stokes parameters, linear birefringence and circular birefringence. This permittivity tensor links the recording interference pattern to the diffraction properties.

Recording gratings with an interference pattern of modulating polarization states require materials, that are sensitive to the polarization state of light instead of the intensity. As it will be shown in Section 5.4, such materials induce directly or indirectly a birefringence modulation instead of just a refractive index modulation.

Finally, in Section 5.5 examples are given of polarization gratings and their diffractive properties are described. A classification of polarization holograms is made based on the interference pattern and formation of birefringence. In Section 5.6 experimental verifications are discussed and initial assumptions on the diffraction theory are evaluated.

### 5.2 Modulating polarization states and birefringence

As described in Chapter 2, the interference pattern is determined by the total electric field of the plane waves. In this section, the focus will be on polarization interference expressed in periodic Stokes parameters. The recorded interference induces birefringence (a polarization hologram). In Section 5.3, this birefringence pattern is described in terms of periodic Stokes parameters in order to describe diffraction. Different types of polarization holograms will be classified.

The interference patterns of polarization holograms can be made explicit with Jones calculus. The Jones components of a polarization grating \(J\) can be written as \(J_{p2} = -J_{s1}^*\) and \(J_{s1} = J_{p2}^*\) for orthogonal polarization states, where \(J^*\) is the complex conjugate of \(J\). Two orthogonal interfering plane waves with wave vectors \(k_1\) and \(k_2\) in the \((x,z)\)-plane with angles \(\pm \theta\) from the z-axis as indicated in Figure 5.1, have an electric field defined by:

\[
E_{\text{holo}} = \sqrt{I_1} \exp(i k_1 \cdot r) \begin{bmatrix} -J_{p}^* \cos \theta \\ J_{s} \\ J_{p} \sin \theta \end{bmatrix} + \sqrt{I_2} \exp(i k_2 \cdot r) \begin{bmatrix} J_{s}^* \cos \theta \\ J_{p}^* \\ J_{s} \sin \theta \end{bmatrix},
\]

\[
\begin{bmatrix} J_{s} \\ J_{p} \end{bmatrix} = \begin{bmatrix} \cos(\psi) \cos(\chi) - i \sin(\psi) \sin(\chi) \\ \sin(\psi) \cos(\chi) + i \cos(\psi) \sin(\chi) \end{bmatrix}.
\]

The intensities \(I_1\) and \(I_2\) of the recording beams are not necessarily equal, allowing for a more general description if compared to previous works [45, 40]. To determine the diffraction of polarization holograms, using the paraxial diffraction theory, it is convenient to use Stokes parameters.

The local Stokes parameters (low-case \(s\)), that describe the interference pattern of a polarization hologram (the recording), are a function of ellipticity angle \(\chi(x)\) and orientation angle \(\psi(x)\). They can be derived by substituting Equation 5.1 into Equation 1.6, resulting in Equation 5.3. In this substitution a small-angle approximation (i.e. \(\sin \theta \approx 0\) and \(\cos \theta \approx 1\)
is used. Therefore, the interference pattern is defined on the $x,y,z$-basis as indicated in Figure 5.1.

$$
\begin{align*}
    s_0(x) &= (I_1 + I_2) \\
    s_1(x) &= (I_1 - I_2) \cos 2\psi \cos 2\chi + 2\sqrt{I_1 I_2}(\cos 2\psi \sin 2\chi \sin \delta(x) - \sin 2\psi \cos \delta(x)) \\
    s_2(x) &= (I_1 - I_2) \sin 2\psi \cos 2\chi + 2\sqrt{I_1 I_2}(\sin 2\psi \sin 2\chi \sin \delta(x) + \cos 2\psi \cos \delta(x)) \\
    s_3(x) &= (I_1 - I_2) \sin 2\chi - 2\sqrt{I_1 I_2} \cos 2\chi \sin \delta(x),
\end{align*}
$$

where $\delta(x) = 2kx \sin \theta$ is a periodic function with period $\Lambda$ along the grating direction $x$. These modulating Stokes parameters are a special case of the general modulating Stokes parameters of Equation 2.8. The recording of these interference patterns of polarization states results in the formation of birefringence. Throughout this thesis, it is assumed that linear polarized light induces linear birefringence and circular polarized light induces circular birefringence [46, 41, 44]. To make a distinction between different Stokes parameters, an upper-case ”$S$” is used to describe the polarization states of the recording light. The polarization selective reading (or diffraction) is described by Stokes parameters with the upper-case ”$S$”. To describe the local birefringence of polarization gratings not only the magnitudes $\Delta n_{lin}$ and $\Delta n_{cir}$ are required but also its local orientation. To describe this orientation of the birefringence, Stokes parameters are used with a lower-case ”$s$”.

Although the interference pattern at a specific instance in time has a period of $2\Lambda$, as shown in Figure 5.2a, the recording of the interference pattern into birefringence leads to a grating with a period of $\Lambda$, as the induced linear birefringence is the same for opposite directions of linear polarized light (Figure 5.2b). The induced birefringence is the time-average interference pattern and has a period as defined by Bragg’s-law, just like the ’classical’ intensity modulations of Section 2.2.1 (Figure 5.2c).

The variations of the Stokes parameters of interference and the birefringence along the grating can be visualized by plotting them on the Poincaré sphere (Section 2.2). Every polarization hologram can be represented on the Poincaré sphere as a circle. Each period $\Lambda$ of the grating in the $x$-direction corresponds exactly to one contour. Depending on the orthogonal polarization states of the recording beams, different circles are described, defined by the normal vector $\mathbf{L}_c$, the center $C_c$ and the radius $r_c$. The normal vector $\mathbf{L}_c$ through $C_c$ is intersecting the origin and points $P_1(s_{11}, s_{21}, s_{31})$ and $P_2(s_{12}, s_{22}, s_{32})$ on the Poincaré sphere representing the polarization states of the recording plane waves. The position of the
Figure 5.2: Periodic modulation of (a) the instantaneous electric field components $E_x$ and $E_y$, (b) the time-average polarization states of an interference pattern of two orthogonal circular polarized plane-waves and (c) an intensity modulation of an intensity hologram.

center $C_c$ on $L_c$ is determined by the intensities $I_1$ and $I_2$ by the following ratio:

$$C_c = L_c \cdot \frac{I_1 - I_2}{I_1 + I_2}.$$  \hspace{1cm} (5.4)

The radius of the circle $r_c$ is defined by:

$$r_c = s_0 \sqrt{1 - \left(\frac{I_1 - I_2}{I_1 + I_2}\right)^2}.$$  \hspace{1cm} (5.5)

These parameters are shown in Figure 5.3. Note, that when the intensity $I_1$ is higher than $I_2$, the circle is closer to $P_1$ than $P_2$. If both intensities of the recording beams are equal ($I_1 = I_2$) the center of the circle $C_c$ is at the origin and the radius $r_c$ equals the radius of the Poincaré sphere.

Every combination of two orthogonal polarization states (of coherent light) can be chosen to create a polarization hologram with a constant intensity, but modulating polarization pattern. Three specific types of polarization holograms, formed with orthogonal linear and orthogonal circular polarized light, respectively, will be described. These polarization holograms will be named after the Stokes parameter that represents the polarization state of the recording beams. For example, a polarization hologram recorded with orthogonal circular polarized light will be referred to as an $\hat{S}_3$ polarization hologram.

$\hat{S}_3$ polarization hologram The interference pattern of right-handed ($\hat{S}_3 = 1$) and left-handed ($\hat{S}_3 = -1$) recording plane waves of equal intensity results in an interference pattern with only linear polarization states as indicated in Figure 5.4a. Therefore, this interference pattern induces only linear birefringence. On the Poincaré sphere this is
Figure 5.3: A circle on the Poincaré sphere describing the modulation of a polarization hologram.

Figure 5.4: Polarization holograms represented as circles on the Poincaré sphere with below them the corresponding interference pattern. a, $\hat{S}_3$ polarization recording with orthogonal circular polarized light. b and c, $\hat{S}_1, \hat{S}_2$ recording respectively with orthogonal linear polarized light (horizontal and vertical for b and at 45 degrees for c). d, arbitrary recording with orthogonal polarized light with different intensity ratios.
represented by a circle in the \((s_1, s_2)\)-plane. In this case \(I_1 = I_2 = 0.5\), \(\chi = \pi/4\) and \(\psi = 0\) and the local Stokes parameters of the birefringence are:

\[
s_1 = \sin \left( \frac{2\pi x}{\Lambda} \right) \quad \text{and} \quad s_2 = \cos \left( \frac{2\pi x}{\Lambda} \right) \quad \text{and} \quad s_3 = 0.
\]

(5.6)

Note that \(2\sqrt{I_1 I_2} = 1\) and the total intensity \(\hat{S}_0\) has been normalized. These Stokes parameters are a function of position \(x\), enabling the calculation of the diffraction efficiencies, as will be shown in Section 5.3.

\(\hat{S}_1\) polarization hologram Using horizontal \((\hat{S}_1 = 1)\) and vertical \((\hat{S}_1 = -1)\) linear polarized light with identical intensities, an interference pattern is created, which is represented by a circle in the \((s_2, s_3)\)-plane on the Poincaré sphere, as indicated in Figure 5.4b. This interference pattern modulates from 45 degrees tilted linear polarized \((s_2 = 1)\) to right handed circular polarized \((s_3 = 1)\), then to -45 degrees tilted linear polarized \((s_2 = -1)\) and finally to left handed circular polarized light \((s_3 = -1)\). This interference pattern, when recorded, results in a corresponding modulation of linear and circular birefringence. Their Stokes parameters modulate as a function of position according to:

\[
s_2 = \cos \left( \frac{2\pi x}{\Lambda} \right) \quad \text{and} \quad s_3 = \sin \left( \frac{2\pi x}{\Lambda} \right) \quad \text{and} \quad s_1 = 0.
\]

(5.7)

\(\hat{S}_2\) polarization hologram The interference pattern of two orthogonal linear polarized plane waves, tilted at 45 degrees \((\hat{S}_2 = -1 \text{ and } \hat{S}_2 = 1)\) looks similar to the previous interference pattern as it modulates from linear to circular to orthogonal linear and orthogonal circular. The representation of the polarization states is a circle on the \(s_1, s_3\) plane and is indicated by Figure 5.4c.

\[
s_1 = \cos \left( \frac{2\pi x}{\Lambda} \right) \quad \text{and} \quad s_3 = \sin \left( \frac{2\pi x}{\Lambda} \right) \quad \text{and} \quad s_2 = 0.
\]

(5.8)

Every polarization hologram recorded by two orthogonal linearly polarized beams results in a modulation of polarization states from circular to linear. The linear state of polarization of the interference pattern is always 45 degrees rotated with respect to the linear polarization state of the recording plane waves. The representation on the Poincaré sphere is a circle through the \(s_3\) axis.

Two orthogonal linearly polarized beams form an interference pattern, which can be represented by a modulation of a Stokes parameter \(s_x = \cos (2\pi x/\Lambda)\) in the \((s_1, s_2)\)-plane and \(s_3 = \sin (2\pi x/\Lambda)\). In Appendix A.1 the general interference pattern of a polarization hologram (Figure 5.4d) is constructed from two arbitrary elliptical orthogonal polarization states.

### 5.3 Diffraction of polarization gratings

Many optical elements that change the intensity and polarization (but not the propagation direction) of light passing through them can be described by a \(2 \times 2\) Jones matrix. Examples of these include wave-plates, polarizers, and optical density filters. It is also well known in paraxial-domain diffractive optics that the diffraction of anisotropic materials can be described...
The parameters describe the modulation (shape and orientation of the ellipse along the grating) and the circular polarized light. Now we come to the important observation, that the Stokes parameter (linear birefringence with linear polarized light and circular birefringence with every Stokes parameter of the dielectric tensor is multiplied by a corresponding birefringence).

By substitution \( n_0 = \sqrt{\varepsilon_0} \), \( \Delta n_{lin} = (\kappa_\parallel - \kappa_\bot) / (2\sqrt{\varepsilon_0}) \) and \( \Delta n_{cir} = \kappa_c / \sqrt{\varepsilon_0} \) the transfer matrix 5.12 is obtained, analog to the general transfer function as described in Section 2.3.1.

\[
T = \exp \left( \frac{-2i\pi d}{\lambda} \right) = \exp \left( \frac{-i\pi d}{\lambda} \left[ \begin{array}{cc} 2n_0 + \Delta n_{lin}s_1 & \Delta n_{lin}s_2 - i\Delta n_{cir}s_3 \\ \Delta n_{lin}s_2 + i\Delta n_{cir}s_3 & 2n_0 - \Delta n_{lin}s_1 \end{array} \right] \right).
\]

The birefringence \((\Delta n)\) has been split into circular and linear birefringence. As mentioned before, it is assumed that the circular birefringence is induced by circular polarized light \((\hat{S}_3)\), and linear birefringence is induced by linear polarized light \((\hat{S}_1\) and \(\hat{S}_2)\). This assumption is justified by the independence of linear and circular polarizations; both components are orthogonal parts of the dielectric tensor [46, 41, 44]. As can be seen from Equation 5.12, every Stokes parameter of the dielectric tensor is multiplied by a corresponding birefringence component (linear birefringence with linear polarized light and circular birefringence with circular polarized light). Now we come to the important observation, that the Stokes parameters describe the modulation (shape and orientation of the ellipse along the grating) and the \(\Delta n\) describes the magnitude of the induced birefringence. The Stokes parameters depend on...
the interference pattern of the light; the magnitude depends on the response of the material to the intensity of the interference pattern. Therefore, Equation 5.12 links the interference pattern to the induced birefringence pattern. Again a lower-case "s" is used to indicate that the Stokes parameter represents the ellipticity of the local birefringence.

For convenience, the transfer matrix is rewritten in a non-exponential form:

\[
T = T_0 \begin{pmatrix}
\cos(F) + iN_{lin} s_1 \sin(F) / F & (iN_{lin} s_2 + N_{circ} s_3) \sin(F) / F \\
(iN_{lin} s_2 - N_{circ} s_3) \cos(F) - iN_{lin} s_1 \sin(F) / F & \cos(F) - iN_{lin} s_1 \sin(F) / F
\end{pmatrix},
\]

(5.13)

with:

\[
F = \sqrt{N_{lin}^2 (s_1^2 + s_2^2) + N_{circ}^2 s_3^2},
\]

\[
T_0 = \exp \left( \frac{-2\pi d n_0}{\lambda} \right),
\]

\[
N_{lin} = \frac{\pi d \Delta n_{lin}}{\lambda},
\]

\[
N_{circ} = \frac{\pi d \Delta n_{circ}}{\lambda}.
\]

The near-field diffraction is obtained by multiplication of the transfer matrix with the incoming electric field vector (a polarized plane wave at normal angles). Analogous to the diffraction of thin isotropic phase gratings (Equation 2.33), the far-field is obtained by applying the Fourier transform of the near field:

\[
D_m = \frac{1}{\Lambda} \int_0^\Lambda T(x)E_{in} \exp \left( \frac{-i2\pi mx}{\Lambda} \right) dx.
\]

(5.14)

For every diffraction order \( m \) a vector \( D_m \) can be calculated representing the electric field in the far field including all polarization information. The angle of the diffracted light \( \theta_m \) of the \( m \)th diffracted order is determined by the grating equation:

\[
\sin \theta_m \cos \phi_{out} = n \sin \theta_{in} \cos \phi_{in} + \frac{m\lambda}{\Lambda}.
\]

(5.15)

For simplicity reasons, the incoming electric field vector is chosen to be independent (constant) of \( x \) along the grating direction. The general solution of the far field diffraction is therefore:

\[
D_m = \begin{pmatrix}
T'_{1,1}(m)E_{in,x} + T'_{1,2}(m)E_{in,y} \\
T'_{2,1}(m)E_{in,x} + T'_{2,2}(m)E_{in,y}
\end{pmatrix}.
\]

(5.16)

Where \( T'_{k,l}(m) \) is the Fourier transform of \( T_{k,l}(x) \) and \( E_{in,x} \) and \( E_{in,y} \) are the \( x \) and \( y \) components of the incoming electric field respectively. Finally, as derived in appendix A.3, the diffraction efficiency \( \eta_m \) is calculated from this general far field and expressed in Stokes parameters resulting in:
\[ \eta_m = |D_m|^2 = \]  \hspace{1cm} (5.17)

\[ \frac{1}{2} \left( |T_{1,1}(m)|^2 + |T_{1,2}^*(m)|^2 + |T_{2,1}(m)|^2 + |T_{2,2}^*(m)|^2 \right) + \]

\[ \frac{1}{2} \left( |T_{1,1}(m)|^2 - |T_{1,2}^*(m)|^2 + |T_{2,1}(m)|^2 - |T_{2,2}^*(m)|^2 \right) \frac{S_1}{S_0} + \]

\[ \frac{1}{2} \left( T_{1,1}(m)T_{1,2}^*(m) + T_{2,1}(m)T_{2,2}^*(m) + T_{1,1}^*(m)T_{2,1}^*(m) + T_{2,1}^*(m)T_{2,2}^*(m) \right) \frac{S_2}{S_0} + \]

\[ - \frac{i}{2} \left( T_{1,1}(m)T_{1,2}^*(m) + T_{2,1}(m)T_{2,2}^*(m) - T_{1,1}^*(m)T_{2,1}^*(m) - T_{2,1}^*(m)T_{2,2}^*(m) \right) \frac{S_3}{S_0}. \]

As mentioned before, the upper-case "S" denotes the Stokes parameters of the reading or diffracting polarized light: the light incident on the polarization grating. These are different from the lower-case "s" used to describe the birefringence of the recorded grating and the upper-case "\( \hat{S} \)" used for the recording polarization states.

Concluding, the polarization selective-diffraction can be determined with Equation 5.17 expressed in Stokes parameters, when the periodic modulations of the circular and linear birefringence with amplitude \( \Delta n_{lin} \) and \( \Delta n_{cir} \) is known along the grating direction. These modulations are determined by the interference pattern of the recording beams and the sensitivity of the material to induce birefringence, when exposed to the interference pattern.

### 5.4 Polarization sensitive materials

Recording the interference pattern of polarization states to form a polarization grating (a modulation of birefringence) requires materials that are sensitive to polarized light. The ability of a material to form birefringence or dichroism is called the Weigert effect and has been reported in literature since the beginning of the 20th century [49, 50]. Mainly two classes of materials are identified: azobenzene polymers [51] and liquid crystals [52]. Both types of materials embody the periodic anisotropy of polarization holograms differently and are addressed separately in this section.

#### 5.4.1 Azobenzene polymers

For azobenzene polymers, the increase of linear and circular birefringence is assumed to be linear for low energy doses, at least above the activation energy. For high exposure doses the induced birefringence becomes independent of intensity. Therefore, the response of anisotropic materials, like azobenzenes [20], can be approximated by [53]:

\[ \Delta n_{lin} = \frac{\Delta n_{l,max}}{1 + (t/\tau_l + tD_lI)^{-1}} \]  \hspace{1cm} (5.18)

\[ \Delta n_{cir} = \frac{\Delta n_{c,max}}{1 + (t/\tau_c + tD_cI)^{-1}}. \]  \hspace{1cm} (5.19)

In these equations \( t \) is the time, \( \tau_l, \tau_c, D_l \) and \( D_c \) are material parameters that can be determined experimentally and \( I \) is the total intensity, which induces the birefringence in the material. It is limited to a maximum linear birefringence \( \Delta n_{l,max} \) and a maximum circular
birefringence $\Delta n_{c,max}$. Initially the response is a linear function of the doses ($I \times t$) and can therefore be further simplified in the case of either very low or very high doses, as will be shown in the examples that follow. These equations link the interference pattern of the polarized light to the material’s birefringence.

Azobenzene polymers induce birefringence by undergoing photo-isomerization cycles when irradiated by linearly polarized light. The azobenzene groups of these polymers are photochromic and anisotropic. They isomerize from trans (or E) to cis (or Z) or visa versa as shown in Figure 5.6. Due to the photo-isomerization, the azobenzene groups change their orientation and tend to line up in a direction perpendicular to the direction of polarization of the excitation as schematically shown in Figure 5.7. When probed with polarized light, such a medium shows an increased absorption and refractive index in the perpendicular polarization direction. Irradiation of the sample with unpolarized or circularly polarized light at normal incidence restores the isotropic absorption.

Photoinduced anisotropy is determined by two quantities: the macroscopic order parameter $\alpha_S$ and the induced birefringence $\Delta n_i$. The macroscopic order parameter is defined as:

$$\alpha_S = \frac{A_\parallel - A_\perp}{A_\parallel + 2A_\perp}, \quad (5.20)$$

where $A_\parallel$ and $A_\perp$ represent the absorbance with a polarization direction parallel and perpendicular to the polarization of the excitation, respectively. The induced birefringence is defined as:
Figure 5.7: Isotropic azobenzene polymer (a) is irradiated by linear polarized light, (b) exciting mainly the azobenzene groups aligned in the direction of the linear polarized light. (c) The reorientation occurs perpendicular to the direction of excitation.

\[ \Delta n_i = n_\parallel - n_\perp, \]  \hspace{1cm} (5.21)

where \( n_\parallel \) and \( n_\perp \) are the refractive indices for polarizations parallel and perpendicular to the polarization of the excitation, respectively.

In contrast to the linear anisotropy, that has been well studied over the last ten years with experimental and theoretical work on both applied and fundamental levels [20], the circular anisotropy has only been recently an active area of research. Photo-induced circular birefringence has been observed in an a-chiral azobenzene-material first by Nikolova et al. [54]. Since then, Nikolovas group have reported studies in which they investigate the induced circular anisotropy for experimental parameters such as the pump lasers polarization [55] and wavelength [56], the type of azobenzene-material [57], and the influence of the circular anisotropy on photo-inscribed diffraction gratings [58].

The mechanism reported by Nikolova of induced circular birefringence is modeled by means of a stepwise propagation of polarized light through a layer. Consider a film of an azobenzene polymer irradiated with circular polarized light, subdivided into an infinite number of thin sublayers. Circular birefringence is induced throughout the thickness of this film. When circularly polarized light passes through a first sublayer of the film, it is transmitted to the second layer as elliptically polarized light. This is caused by the interaction with the linear optical anisotropy of the sublayer. The interactions in the second layer will have a similar effect on the polarization state of the irradiated polarized light and therefore change the azimuth of the polarization sent to the next layer. This yields a self-rotation of the light by the excited azobenzene groups and visa versa.

### 5.4.2 Liquid crystal materials

As mentioned in Section 3.3, liquid crystals have multiple refractive indices, which arise from their molecular shape (rod or disk-like). To induce a macroscopic birefringence, the liquid crystals need to be oriented. In general they can be oriented by means of an orientation layer,
an electric field or a flow, but for polarization holograms only an orientation layer is feasible.

Photo-alignment films (referred to as linear-photo-polymers, or LPP [59, 35, 60, 61, 62]), can be coated on a substrate and are sensitive to linear polarized light. Figure 5.8 shows the schematic recording of the orientation of the local polarization state of an interference pattern in LPP. Two orthogonal circular polarized plane waves form an interference pattern as indicated in Figure 5.4a. A linear birefringence results after coating a liquid crystal layer onto the LPP alignment layer. No circular birefringence is induced ($\Delta n_{\text{cir}} = 0$). The linear birefringence originates from the difference between the ordinary and extra-ordinary refractive index of the liquid crystals and is independent of the exposure dose of the holographic recording in the LPP layer. The magnitude of birefringence remains constant over the exposed area and is determined by the degree the liquid crystal is oriented. Therefore, the value of $\Delta n_{\text{lin}}$ can be considered constant and Equation 5.6 applies, as verified experimentally by Escuti and coworkers [60].

In the field of liquid crystal materials, the property of circular birefringence is well known for its application in LCDs. Adding a chiral molecule to a liquid crystal medium induces a helical structure known as the cholesteric phase [63] by self-assembly. Such macroscopic helical structures exhibit circular birefringences up to four orders of magnitude larger than those obtained by azobenzene polymers. Despite the high circular anisotropy, cholesteric liquid crystals are not well suited for optical recording of polarization gratings. This is due to the self-assembly of liquid crystals, making it difficult to create modulations of right-handed to left-handed helical structures alternating with linear birefringent modulations, with small periods [11].

A circular birefringent material, that could be used for polarization gratings, is a light-driven molecular rotor [64]. The molecular structure of this type of material is indicated in Figure 5.9. Starting with a left-handed circular birefringent isomer, the molecular rotor switches to the right-handed isomer upon UV-exposure. The isomerization as a function of UV-exposure has been monitored by measuring the optical rotation angle, as indicated in Figure 5.10a. The optical rotation of a medium is a result of circular birefringence as indicated by:

$$\alpha = \alpha_0 \frac{j}{\lambda_0} \Delta n_{\text{cir}}, \quad (5.22)$$
with $\alpha$ the optical rotation, $\alpha_0$ an experimentally determined material parameter, $l$ the optical path length and $\lambda_0$ the wavelength of measurement. First the isomerization responds as a linear function of intensity, just as it is assumed in the theoretical model. For higher exposure doses the conversion slows down and decays exponentially. The right-handed circular birefringent isomer can be switched back (isomerized) by heating. The isomerization as a function of heating time at 60°C has been measured as well and is indicated in Figure 5.10b.

### 5.5 Examples of polarization holograms

In the previous sections a theoretical framework has been presented describing the formation of interference patterns of modulating polarization states, the modulation of linear and circular birefringence, the recording mechanism by polarization sensitive materials, and the diffractive properties of polarization gratings. In the next sections, several examples of polarization gratings are given. Each example demonstrates the formation of a specific polarization-selective diffraction for a certain polarization sensitive material and interference pattern. As such, polarization holograms are described, that are formed by interference patterns of circular polarized light, the so called $\hat{S}_3$-polarization hologram, and holograms that are formed by interference patterns of linear polarized light, the so called $\hat{S}_1$- and $\hat{S}_2$-polarization holograms. In every section the diffractive properties are determined for materials with different
anisotropic responses as schematically depicted in Figure 5.11.

5.5.1 \(S_3\)-polarization hologram with \(\Delta n_{lin}, \Delta n_{cir} \neq 0\)

In this first example it is assumed that the material is able to form both linear and circular birefringence, without constraints. The linear birefringence \(\Delta n_{lin}\) and circular birefringence \(\Delta n_{cir}\) scales linear with the intensity upon exposure to linear and circular polarized light, respectively. Two circular orthogonal polarized plane waves of equal intensity form an interference pattern with only linear polarization states as indicated in Section 5.2. As the ellipticity angle in the interference pattern is zero, no circular birefringent modulation is induced. However, a constant initial circular birefringence may be present within the material (expressed by \(s_3 = A_c\)). Reasons for a non-zero \(A_c\) may be the presence of an optical active compound (not induced by interfering light) or a difference in recording plane wave intensities \(I_1\) and \(I_2\). In the last case the \(s_3(x)\) as derived in Equation 5.3 is not equal to zero. This simplifies the general transfer matrix 5.13 to:

\[
\begin{align*}
T_{1,1} &= T_0 \left( \cos(N_{bir}) + i \frac{N_{lin} \sin(N_{bir})}{N_{bir}} \cos \left( \frac{2\pi x}{\Lambda} \right) \right) \\
T_{1,2} &= T_0 \left( \frac{\sin(N_{bir})}{N_{bir}} \left( i N_{lin} \sin \left( \frac{2\pi x}{\Lambda} \right) + N_{cir} A_c \right) \right) \\
T_{2,1} &= T_0 \left( \sin(N_{bir}) \left( i N_{lin} \sin \left( \frac{2\pi x}{\Lambda} \right) - N_{cir} A_c \right) \right) \\
T_{2,2} &= T_0 \left( \cos(N_{bir}) - i \frac{N_{lin} \sin(N_{bir})}{N_{bir}} \cos \left( \frac{2\pi x}{\Lambda} \right) \right),
\end{align*}
\]

with:

\[
N_{bir} = \sqrt{N_{lin}^2 + N_{cir}^2 A_c^2}.
\]

To determine the far field (Equation 5.14), the Fourier transform is taken from each element of the matrix for every diffraction order \(m\). This results in non-zero values only for the 0th and 1st diffraction orders as:
D_0 = T_0 \left[ \cos(N_{br})E_{in,x} + N_{cir}A_c \text{sinc}(N_{br})E_{in,y} \
- N_{cir}A_c \text{sinc}(N_{br})E_{in,x} + \cos(N_{br})E_{in,y} \right] \quad (5.25)

D_{\pm 1} = \frac{1}{2} T_0 N_{lin} \text{sinc}(N_{br}) \left[ iE_{in,x} + E_{in,y} \\ E_{in,x} - iE_{in,y} \right].

The polarization dependent diffraction can be determined by calculating the diffraction efficiencies of both orders with Equation 5.17.

\eta_0 = \frac{\Delta n_{lin}^2 A_c^2}{\Delta n_{cir}^2 + \Delta n_{lin}^2} \sin^2(N_{lin} + N_{cir}A_c) + \cos^2(N_{lin} + N_{cir}A_c) \quad (5.26)

\eta_{\pm 1} = \frac{1}{2} \frac{\Delta n_{lin}^2}{\Delta n_{circ}^2 A_c^2 + \Delta n_{lin}^2} \sin^2(N_{lin} + N_{cir}A_c) \left( 1 \mp \frac{S_3^2}{S_0^2} \right).

Examining the diffraction properties of this polarization grating, the following characteristics are obtained:

- The diffraction efficiency \( \eta \) of the 0th order is not a function of the Stokes parameters, meaning it is independent of the polarization state of the incoming light.
- The first order of diffraction depends on \( S_3 \), the amount of circular polarized light.
- In case of linear polarized light (\( S_3 = 0 \)), the diffraction efficiency is equal for both first orders.
- Right-handed polarized light is diffracted in the 1st order and left-handed polarized light is diffracted from the -1st order.

These polarization dependent diffraction efficiencies are illustrated in Figure 5.12. The diffraction efficiency is plotted for \( \eta_0 \) and \( \eta_1 \). For \( \eta_{-1} \) the results are identical to \( \eta_1 \) for Figure 5.12a and mirrored around \( S_3 = 0 \) for Figure 5.12b. As can be observed, circular birefringence decreases the diffraction efficiency in an \( S_3 \)-polarization grating.

In Figure 5.12a the diffraction efficiency of the first order shows a decrease for an increasing magnitude of circular birefringence. Practical values of \( N_{cir} \cdot A_c \) have an upper limit of approximately 1, either due to a limited layer thickness or obtainable \( \Delta n_{cir} \). For low induced linear birefringence (i.e. \( N_{lin} < 0.5\pi \)) the diffraction efficiency has an optimum at \( N_{cir} \cdot A_c > 0 \).

In Figure 5.12b the diffraction efficiency as a function of the intensity in terms of \( S_3 \) is shown. As can be seen the 0th diffraction order is constant and \( N_{lin} \) determines its value. For the first diffractive order there is a linear dependence on the intensity of circular polarized light. For right-handed polarized light (\( S_3 = 1 \)) the diffraction efficiency of the 1st order is always zero. For left-handed polarized light the diffraction efficiency is always zero for -1st order.

Summarizing, it is possible to control the diffraction efficiency of circular polarized light using polarization holograms induced by the interference of circular polarized light. Right-handed and left-handed polarized light are split to the minus first and first diffraction orders respectively. The total diffraction efficiency (the sum of the diffraction efficiencies of both first orders) is determined by the \( \Delta n_{lin} \) and thickness of the grating. The intensity ratio between the two first orders is determined by the polarization state of the incoming light.
5.5.2 \( \hat{S}_3 \)-polarization hologram with \( \Delta n_{cir} = 0 \)

If there is no circular birefringence present in the material \( (A_c = 0) \) the diffraction efficiencies simplify to Equation 5.27. The zeroth order, depending only on layer thickness, linear birefringence and wavelength is independent of the polarization of light. A 100\% diffraction efficiency for purely circular polarized light is obtained for the first orders of diffraction if the grating has \( N_{lin} = \pi/2 \), i.e. \( d\Delta n_{lin} = \lambda/2 \). This corresponds to the points on the vertical axis of Figure 5.12a and to the solid black line in Figure 5.12b. In this case left-handed circular polarized light is diffracted in the 1st order of diffraction with 100\% efficiency and the 0th and -1st order of diffraction have consequently 0\% diffraction efficiency. Right-handed circular polarized light is diffracted with 100\% diffraction efficiency in the -1st order:

\[
\eta_0 = \cos^2(N_{lin}) \\
\eta_{\pm 1} = \frac{1}{2} \left( 1 \mp \frac{S_3}{S_0} \right) \sin^2(N_{lin}).
\]

This is the special case of the example in Section 5.5.1 that give the highest diffraction efficiencies. High diffraction efficiencies are desirable for optical switches and display applications. This example has been experimentally verified, and will be discussed here.

Similar to the work of Escuti [65] \( \hat{S}_3 \)-polarization gratings without circular birefringence have been made using linear photo-alignment polymer (LPP) CP03 of Rolic. First, a layer of LPP with a thickness of 50 nm is spin-coated on a glass substrate. After baking of the solvent (15 min at 90\°), the LPP is exposed to an interference pattern of right- and left-handed polarized light of an Argon UV-laser of 351 nm with a dose of 5.7 mW/cm\(^2\). Due to this exposure the interference pattern of modulating linear polarization states is captured in the form of an alignment pattern by the LPP. After spin-coating a layer of a reactive nematic liquid crystals mixture (RMs06 of Merck) on the LPP alignment layer, the solvent is backed off at 50\°C. The director of the liquid crystals follows the alignment pattern and
therefore a polarization hologram is obtained. Finally, the polarization grating is fixated by a photo-polymerization (a UV-exposure) in a nitrogen atmosphere for 8 minutes. By choosing different spin speeds the layer thickness of the liquid crystals can be controlled.

The polarization-selective diffraction of an $\hat{S}_3$-polarization grating with a period $\Lambda$ of 8.0 $\mu$m and thickness $d$ of 0.41 $\mu$m has been analyzed by a HeNe-laser at normal angle. The intensities of the $\pm 1$ diffraction orders were measured, while changing the polarization state of the laser from left-handed circular to linear and to right-handed circular by rotating a quarter wave-plate as indicated in Figure 5.13a.

Figure 5.13b shows the relative intensities of the diffractive orders. At position 1, right-handed circular polarized light is diffracted towards the -1st order and no diffraction occurs in the +1st order. If the quarter wave-plate is rotated to 45 degrees, indicated by position 2, the incoming light is linear polarized. It is diffracted in equal intensities of right- and left-handed circular polarized light in their corresponding diffraction orders. At position 3, left-handed circular polarized light is diffracted towards the +1st order and no light is diffracted in the -1st order. The 0th order has always the same polarization state as the incoming laser.

As predicted by the theoretical framework, the $\hat{S}_3$-polarization gratings show polarization-selective diffraction of circular polarized light.

5.5.3 $\hat{S}_1$- and $\hat{S}_2$-polarization holograms with $\Delta n_{\text{lin}}, \Delta n_{\text{cir}} \neq 0$

In this example we will use two arbitrary, orthogonal, linearly polarized plane waves for the recording. Recall, polarization gratings created with two orthogonal linear polarized recording plane waves represent a circle on the Poincaré sphere as illustrated in Figure 5.4b (for horizontal and vertical linear polarized recording beams) and 5.4c (for orthogonal at 45 degrees). In general, a circle through the $s_3$-axis on the Poincaré sphere represents an interference pattern of two orthogonal linear polarization states. Therefore, the interference
pattern will always include right and left-handed polarized states.

The grating, formed after exposure, modulates between linear and circular orthogonal states of birefringence (orthogonal in direction or handedness); $s_1^2 + s_2^2 = \cos(2\pi x/\Lambda)$ and $s_3 = \sin(2\pi x/\Lambda)$. The corresponding transfer matrix, derived from Equation 5.13 is:

$$T = T_0 \begin{bmatrix} \cos(F) + i \frac{s_N}{\sqrt{1 + \rho^2}} \sin(\frac{2\pi x}{\Lambda}) & i \frac{s_N}{\sqrt{1 + \rho^2}} \cos(\frac{2\pi x}{\Lambda}) \\ i \frac{s_N}{\sqrt{1 + \rho^2}} \sin(\frac{2\pi x}{\Lambda}) - N_{cir} \sin(N_{lin}) \sin(f) & \cos(F) - i \frac{s_N}{\sqrt{1 + \rho^2}} \cos(\frac{2\pi x}{\Lambda}) \sin(f) \end{bmatrix}, \quad (5.28)$$

where $\rho$ is the ratio $s_1/s_2$ indicating the orientation of the linear birefringence. Note that $\rho$ also equals $\tan(2\psi)$. Therefore, if $\rho$ equals zero the linear birefringence direction is oriented in the $s_1$ direction. If $\rho$ equals infinity it corresponds to the $s_2$ direction. For every grating recorded with orthogonal linear polarized light, there is a value of $\rho$ between zero and infinity ($0 \leq \arctan(\rho) \leq \pi/2$).

To determine the polarization dependent diffraction efficiency each element of this transfer matrix has to be integrated over $x$. A third order Fourier approximation is made, as shown in Appendix A.4, in order to find an analytical solution. The higher order Fourier terms stay below 0.01 if both $N_{lin}$ and $N_{cir}$ are less than 1, and therefore are assumed negligible. Also for many other values of $N_{lin}$ and $N_{cir}$ this approximation is valid. For $N_{lin} = N_{cir}$ this relation is exact. It has been checked for the cases that will be presented in Figure 5.14 and the error is never larger than a few percent. Integration of the transfer matrix over $x$ together with Equation 5.17 yields the following polarization dependent diffraction efficiencies:

$$\eta_0 = \frac{1}{4} (\cos(N_{lin}) + \cos(N_{cir}))^2$$

$$\eta_{\pm 1} = \frac{1}{64} \left( 9 + \frac{\Delta n_{lin}^2}{\Delta n_{cir}^2} \right) \sin^2(N_{cir}) + 6 \left( N_{cir}^2 + N_{lin}^2 \right) \sin(N_{cir}) \sin(N_{lin}) +$$

$$\left( \frac{\Delta n_{cir}^2}{\Delta n_{lin}^2} + 9 \right) \sin^2(N_{lin}) \pm \frac{N_{cir} N_{lin}}{32} (3 \sin(N_{cir}) + \sin(N_{lin})) \cdot$$

$$\sin(N_{cir}) + 3 \sin(N_{lin}) \left( \frac{S_1 - \rho S_2}{S_0 \sqrt{1 + \rho^2}} \right)$$

$$\eta_{\pm 2} = \frac{1}{16} (\cos(N_{lin}) - \cos(N_{cir}))^2$$

$$\eta_{\pm 3} = \frac{1}{64} (\sin(N_{lin}) - \sin(N_{cir}))^2 \left( \frac{N_{cir}^2 + N_{lin}^2}{2 N_{cir} N_{lin}} \pm 2 N_{cir} N_{lin} \left( \frac{S_1 + \rho S_2}{S_0 \sqrt{1 + \rho^2}} \right) \right).$$

The 0th and $\pm 2$nd orders of diffraction are independent of the polarization state, just like in $\hat{S}_3$-polarization holograms. For the $\pm 1$st and $\pm 3$rd diffraction orders, the diffraction efficiency depends on the polarization state of recording. Note that when both induced (linear and circular) birefringences have the same magnitude, the $\pm 2$nd and $\pm 3$rd diffraction orders disappear and 100% diffraction efficiency of the first order can be obtained.

Figure 5.14a illustrates the diffraction efficiency as a function of linear polarized light of the reading beam with a specific orientation. The orientation varies between the two orthogonal linear polarization recording states. Only $\eta_0$ and $\eta_1$ are shown, as $\eta_{-1}$ is again the mirror image of $\eta_1$. The $\eta_2$ and $\eta_3$ are not shown as they are orders of magnitude smaller. Furthermore, $\eta_0$ is zero for $N_{lin} = N_{cir} = 0.5\pi$ for all orientations and is therefore also not
Polarization holography

Figure 5.14: Diffraction efficiency of the 0th and 1st order for different ratios of $N_{cir}/N_{cir}$ with $N_{cir} = \frac{1}{2} \pi$ (a) as a function the the recorded state of linear polarized light ($\hat{S}_1 + \rho \hat{S}_2$) and (b) as a function of $\rho$ for $\hat{S}_1$ linear polarized light.

plotted. The diffraction efficiency of the first order, $\eta_1$, depends linear on the orientation of linear polarized light. A diffraction efficiency of 100% can be obtained for a ratio $N_{lin}/N_{cir}$ of 1.

Analogous to the example from Section 5.5.1, the $\hat{S}_1$- and $\hat{S}_2$-polarization gratings diffract linear polarized light. The grating essentially splits light into four orthogonal linear polarized diffraction orders with an orientation corresponding to the recording polarization states. $\hat{S}_1$-polarization gratings diffract $S_1$ polarized light selectively. $\hat{S}_2$-polarization gratings diffract $S_2$ polarized light selectively. In general, linear polarization gratings are polarization-selective to their linear polarization recording state.

Figure 5.14b shows the diffraction efficiency of an $\hat{S}_1$-polarization grating as a function of recorded linear polarized light orientation arctan($\rho$). The diffraction efficiency of the first order has its maximum for $\rho = 0$, corresponding to one of the recording polarization states. If the orientation of an incoming linear plane wave is 45 degrees rotated from the orientation of the recording plane waves, the diffraction efficiency of the first diffraction orders have equal intensity (for $\rho = \infty$).

The diffraction efficiency depends on the ratio of linear and circular birefringence. The highest diffraction efficiency is obtained at a ratio of 1. Diffraction efficiencies for a ratio $N_{lin}/N_{cir} = 1$ and $N_{lin}/N_{cir} = 1.5$ are indicated in both graphs of Figure 5.14 as an example. For large deviation from a ratio of 1 the third order approximation is not always sufficient and higher order term should be taken into account. In the next sections, the diffraction efficiency is shown for examples of polarization gratings, with no circular or linear birefringence.

5.5.4 $\hat{S}_1$ and $\hat{S}_2$-polarization holograms with $\Delta n_{lin} = 0$

In this example polarization gratings with only circular birefringence modulation are discussed, which are induced by an $\hat{S}_1$- or $\hat{S}_2$-polarization interference pattern. The modulation of linear birefringence is assumed to be zero, e.g. due to material properties. The grating modulates in this case from right-handed circular to left-handed circular birefringent and is isotropic in between as schematically indicated in Figure 5.15.
Chapter 5

Figure 5.15: Modulation of the circular birefringence of a $\hat{S}_1$- or $\hat{S}_2$-polarization grating

Figure 5.16: Modulation of the linear birefringence of an $\hat{S}_1$-polarization grating

The transfer matrix is simplified in this case to:

$$ T = T_0 \begin{bmatrix} \cos \left( N_{\text{cir}} \cos \left( \frac{2\pi x}{\Lambda} \right) \right) & \sin \left( N_{\text{cir}} \sin \left( \frac{2\pi x}{\Lambda} \right) \right) \\ -\sin \left( N_{\text{cir}} \sin \left( \frac{2\pi x}{\Lambda} \right) \right) & \cos \left( N_{\text{cir}} \sin \left( \frac{2\pi x}{\Lambda} \right) \right) \end{bmatrix}. \quad (5.30) $$

Just like in the previous examples the Fourier transform (integral) is taken and the following diffraction efficiencies are obtained.

$$ \eta_0 = J_0^2(N_{\text{cir}}) \quad (5.31) $$

$$ \eta_1 = J_1^2(N_{\text{cir}}) $$

$$ \eta_2 = J_2^2(N_{\text{cir}}) $$

... 

where $J_n$ are Bessel functions of the first kind with order $n$. There is no polarization-selectivity, as all the diffraction efficiencies are independent of the Stokes parameters. The diffraction efficiency as a function of the material parameters ($N_{\text{cir}}$) is a Bessel function ($J_n$), similar to isotropic phase gratings with a sinusoidal refractive index modulation. This type of grating is therefore not considered to be a polarization grating.

5.5.5 $\hat{S}_1$- and $\hat{S}_2$-polarization holograms with $\Delta n_{\text{cir}} = 0$

In this final example, an $\hat{S}_1$-polarization grating is used to determine the diffractive properties of a polarization sensitive film that only induces a modulation of linear birefringence as indicated in Figure 5.16.

$$ T = T_0 \begin{bmatrix} \cos \left( N_{\text{lin}} \cos \left( \frac{2\pi x}{\Lambda} \right) \right) & -\sin \left( N_{\text{lin}} \cos \left( \frac{2\pi x}{\Lambda} \right) \right) \\ \sin \left( N_{\text{lin}} \cos \left( \frac{2\pi x}{\Lambda} \right) \right) & \cos \left( N_{\text{lin}} \cos \left( \frac{2\pi x}{\Lambda} \right) \right) \end{bmatrix}. \quad (5.32) $$

The polarization diffraction efficiency is determined in two steps. First taking the Fourier transform, then applying Equation 5.17 yields the following diffraction efficiencies:
Polarization holography

Figure 5.17: Modulation of the linear birefringence of an $\hat{S}_2$-polarization grating

\[
\eta_0 = J_0^2(N_{lin}) \\
\eta_1 = J_1^2(N_{lin}) \\
\eta_2 = J_2^2(N_{lin}) \\
\ldots
\]

Similar to the previous example, the diffraction efficiencies are polarization independent and are Bessel functions. For linear birefringent modulation a diffraction is obtained for every order. This kind of polarization gratings is very similar to isotropic phase gratings; the only difference is the birefringence modulates instead of the refractive index.

If an $\hat{S}_2$-polarization grating would have been chosen, a different modulation and transfer matrix results as indicated by Figure 5.17 and Equation 5.34:

\[
T = T_0 \begin{bmatrix}
\exp(iN_{lin} \cos(\frac{2\pi x}{\Lambda})) & 0 \\
0 & \exp(-iN_{lin} \cos(\frac{2\pi x}{\Lambda}))
\end{bmatrix}.
\]

Although the transfer matrix is different, exactly the same diffraction efficiencies would be obtained. As such these gratings have similar diffractive properties as isotropic thin phase gratings as discussed in Chapter 2.

5.6 Conclusions and discussion

Polarization holography is a powerful tool to make anisotropic gratings. Interference of modulating polarization states of light instead of intensities allows the production of gratings with modulating linear and circular birefringence. By using polarization sensitive materials like azobenzene-polymers and LPP orientation layers, the polarization modulation can be recorded.

A theoretical framework has been presented to predict the diffractive efficiencies of anisotropic gratings. High polarization-selectivity is predicted as well as a high diffraction efficiency. Contrary to thin isotropic gratings, thin polarization gratings are able to diffract with 100% efficiency. The theoretical framework is used to design the polarization-selectivity of diffraction. It was shown the polarization state of diffraction corresponds to the polarization state of recording. Only when the magnitude of linear and circular birefringence are the same in magnitude as the linear and circular polarization states of the interference pattern, a 100% diffraction efficiency can be achieved.

To illustrate the applicability of the theoretical framework, several examples have been presented, revealing the polarization-selectivity of diffraction of polarization holograms. Every example assumed that linear polarized light induces linear birefringence and that circular polarized light induces circular birefringence. Furthermore, the magnitude of birefringence is
induced as a linear function of intensity or that after recording it follows the modulation of the interference pattern. The polarization-selectivity and the diffraction efficiency have been calculated for each example.

Further assumptions and predictions that have implicitly been made in the theoretical framework and the described examples deserve some elaboration and will be discussed here.

The birefringence has been split in a linear and circular part. Jones mentions in Ref [7] the possibility that materials exist with a further split of the linear birefringence for linearly polarized light parallel (S1) and at 45° (S2) to the optical axis (a birefringence for every Stokes parameter). In this framework this split is unnecessary as birefringence is induced. Starting with an initially isotropic material, the linear birefringence is modeled by a single Δnlin, that depends on the amount of linearly polarized light $\sqrt{(S_1)^2 + (S_2)^2}$ of the interference pattern.

In Section 5.5.3 a third order Fourier approximation has been made to analytically integrate the transfer matrix of the S1- and S2-polarization holograms. This approximation is valid for ratios of $N_{lin}/N_{cir}$ close to 1. For large deviations higher order Fourier approximations are needed and consequently more diffraction orders appear. For a ratio of 0 or $\infty$ the Bessel functions are obtained as shown in Section 5.5.4 and Section 5.5.5 respectively.

Furthermore, the assumption is made that the material in which the hologram is recorded is so thin that depth variations of the recording field can be neglected. This is also always assumed for thin gratings. This assumption also implies that there is no angular dependency of the diffraction. As such the theoretical framework does not apply to slanted gratings described in Chapter 3.

To model thick polarization gratings a new theory needs to be derived, either analytically or numerically. For an analytical coupled wave theory the description of a continuous change of the polarization states of the propagation wave through the grating will be required (for linear birefringent polarization holograms this has been done by Huang and Wagner [48]). For a numerical approach, like a finite element method, a description of a single period of the polarization grating in terms of the dielectric tensor could be used.

Linear polarization-selective diffraction gratings require the modulation of linear to circular birefringence. To obtain a 100% diffraction efficiency in only a first order of diffraction, linear and circular birefringence should be equal in magnitude. Only theoretical predictions of the diffractive properties have been shown for such birefringent gratings. To verify these gratings experimentally, the molecular rotor, described in Section 5.4, could be used. Creating a holographic film with such a rotor, should be researched in order to obtain a polarization gratings with linear and circular modulating birefringence. A possible route would be to mix a left-handed molecular rotor with a nematic LC mixture and to coat it on an aligned LPP layer. After coating, an exposure to an intensity interference pattern with the same period as the LPP layer isomerizes the molecular rotor locally. Careful attention should be taken to the concentration of the molecular rotor in order to match the magnitude of the circular and linear birefringence. Furtermore, the isomerization should be able to yield equal right-handed and left-handed circular birefringence.
Chapter 6

Micro-structuring of functional layers in OLEDs

Out-coupling structures, that improve the efficiency of display systems, are not limited to LCD applications. They can also be applied to Organic Light-Emitting Diode (OLED) based displays. Although polarization-selective emission is not required for OLED displays, gratings can still be used to improve their efficiency. A lot of light is trapped inside the OLED due to total internal reflection, coupling this light out, increases the total amount of emitted light and therefore the efficiency.

An OLED is a special type of light-emitting diode, in which the emissive layer comprises a thin polymer film, such as polyarylenes, poly(p-phenylene vinylene) and polyheterocycles. OLEDs have their applications mainly in the field of (large area) light-emitting devices. OLED based display devices can be advantageous, if ease of manufacturing is important. In general, OLED displays are cheaper than LCDs [66]. However, the degradation of OLED materials thus far limits their lifetime and is a topic of research [67]. Increasing the efficiency of an OLED also increases the life-time, as a lower current is required for the same light intensity.

An OLED works on the principle of electroluminescence. It is build up of a stack of layers supported on a glass or plastic substrate and, typically, sealed by an aluminium cap to prevent intrusion of water [68], as shown in Figure 6.1a. The first layer of this stack is a transparent anode, that creates an electron deficit, when a current flows through the device. Indium tin oxide (ITO) is commonly used. As a second layer, a conducting polymer (like Poly(3,4-ethylenedioxythiophene) (PEDOT) [69]) transports the "holes" from the anode. The third layer, a polymer luminophore, emits light while transporting electrons from the cathode. Derivatives of PPV, poly(p-phenylene vinylene) and poly(fluorene) are most commonly used as polymer luminophores in OLEDs [70]. The key to the operation of an OLED is the generation of excitons inside this layer. These excitons consists of a bound excited electron and hole pair. When the exciton’s electron and hole combine, a photon is emitted.

OLEDs are inefficient in the sense that a lot of light, that is generated by the polymer luminophores, never reaches the viewer. Internally OLEDs are very efficient (up to 99% quantum yield); externally OLED light extraction efficiency is limited to about 7% (approximated by $1/(4n_{ITO}^2)$) due to the high refractive indices of the inorganic and organic layers (1.7-1.9) and the glass substrate ($n_{glass} = 1.5$). Light, generated in the PPV layer, gets totally internally reflected at the ITO-glass interface (Figure 6.1b position 2), at the glass-air interface (Figure 6.1b position 1) or gets transmitted. The transmission cone is indicated by
Increased light extraction (the half-angle $\gamma$) would enable a higher efficiency, brightness and lifetime. In literature many methods can be found to increase the extraction efficiency of OLEDs. To overcome the total internal reflection from the glass-air interface, Forrest et al. [71] obtained a efficiency gain of 1.5 by putting an micro-lens array on top of the glass substrate. The micro-lenses primarily widen the escape cone for the totally internal reflected light incident at the glass-air interface. Schnitzer et al. [72] report a 30% increase in efficiency by roughening the top side of the glass. Small random structures in the surface scatter light randomly, which is otherwise internally reflected.

Lee et al. [73] and Boroditsky et al. [74] address the light extraction from the organic-ITO interface by introducing a 2D photonic crystal between the glass and ITO layer. A high refractive index material (Silicon Nitride) with a periodic structure ranging from 200 to 800 nm converts waveguided waves into external leaky waves. An experimental increase of 50% efficiency has been observed with this method. An increase of 25% at high energy levels in efficiency has been obtained by McGehee et al. [75] by putting a Bragg grating in the ITO layer to scatter the waveguided light out.

In this chapter, another method of light extraction from OLEDs is investigated. Gratings are created in the functional layers of OLEDs, similar to the work of Friend and Huck [76], where a 2D pattern was fabricated inside the light emitting polymer by inject-printing. Instead of using inject-printing, the gratings in this research are created by pulsed holography. Potentially, the light extraction is improved, since light is diffracted instead of total internally reflected. In the sections below, gratings have been made in several layers of yellow Philips OLEDs. The emission spectrum of these OLED is shown in Figure 6.1b. After structuring one of the functional layers, the OLED was constructed and the angular light distribution and electrical properties were measured to verify that the OLEDs are operational.

These OLEDs consist of an ITO layer ($n_0$ of 1.795 at 600 nm) on a glass substrates coated with PEDOT ($n_0$ of 1.502 at 600 nm) and a PPV derivative (from hereon referred to as PPV, with an average refractive index $n_0$ of 1.865 at 600 nm). In Section 6.1, gratings have been made in the Poly(3,4-ethylenedioxythiophene) (PEDOT) layer and the optical angular and

Figure 6.1: (a) OLED build up of a stack of layers showing the escape cone with half-angle $\gamma$, TIR of the 1. glass-air interface and 2. ITO-glass interface. (b) Spectrum of a Philips yellow OLED.
6.1 Structuring the PEDOT layer

6.1.1 Experiment

Poly(3,4-ethylenedioxythiophene) (PEDOT) was spin coated on an ITO glass substrate with a layer thickness of 100 nm. To induce a grating, a single 4 ns pulse from a Nd:YAG laser of 532 nm is used. Powers up to 172 mJ/cm$^2$ were generated. As mentioned before, a two-beam holographic setup was used with an intensity interference pattern with a period of 800 and 2000 nm.

To determine the sensitivity of the PEDOT to the laser’s energy a 100 nm thick PEDOT layer on an glass-ITO substrate is exposed to an energy level ranging from 12 - 172 mJ/cm$^2$. After exposing (and structuring) the PEDOT, a layer of a PPV is coated onto the OLED and a barium and aluminum cathode is evaporated on it. Finally, after capsulation an OLED is obtained. Multiple OLEDs where placed on one single substrate in such a way, that it consists of 18 exposed OLEDs and 6 reference samples, which could be addressed individually.

By applying a voltage on the OLEDs, light is emitted. The optical characterization of the angular emission is done with a DMS 703 (Autronic-MELCHERS GmbH, Germany) equipped with a diode array spectrophotometer. An I-V curve is obtained by applying a voltage and
measuring the corresponding current. For every voltage, the intensity is measured at normal angle for 550 nm, and a current-intensity (I-L) curve is obtained.

### 6.1.2 Results and discussion

A large variation in the intensity (estimated at 25%) of the emission at the normal was observed between reference samples of different OLED substrates. However, some trends could be observed between the OLEDs from the same substrate. PEDOT layers, exposed with a total energy of 12-56 mJ/cm$^2$, did not show any grating structure and did not diffract light. After constructing an OLED and applying a voltage, they did not show significant changes in light intensity or angular distribution compared to the non-exposed reference samples.

Exposing PEDOT to an energy ranging from 56 to 91 mJ/cm$^2$ resulted in clearly visible and diffracting gratings. Surface analysis by scanning electron microscopy (SEM) confirmed the existences of these gratings as indicated in Figure 6.3. A trend was observed when increasing the energy. More PEDOT is removed with higher energy doses and wider and deeper grooves are generated. Exposures to higher energy doses than 100 mJ/cm$^2$ result in holes and wide gaps in the structure. Too much PEDOT was removed in these cases, as indicated by the interferometry pictures of Figure 6.4a. The OLEDs made from these samples

![Figure 6.3: SEM analysis of structured PEDOT with a range of exposure intensities (top-view)](image)
Micro-structuring of functional layers in OLEDs

Figure 6.4: a.) Surface topology of a structured PEDOT layer with a pulse of about 172 mJ/cm² showing gaps and cracks in the grating. b.) Angular light distribution from an OLED with a structured PEDOT layer (exposed to 91 mJ/cm²) with a periodicity Λ of 800 nm operated at 8V.

did not show any light output.

The emission spectrum of the yellow Philips OLEDs, as seen in Figure 6.1b, has a peak intensity around 550 nm. Therefore, the angular measurements are done at this wavelength. The detection angle Ψ is measured from the normal and compared to a reference sample, to analyse if the angular output changed.

As can be seen from Figure 6.4b, the angular intensity distribution of an unstructured OLED (reference sample) is close almost constant (= intensity does not change with angle). The structured OLED with a grating period of 800 nm do not show a significant change in angular distribution operated at 8V. Some structured OLEDs showed an huge increase at higher angles due to light leakage at the borders of the OLEDs, especially for the gratings with a period of 2000 nm.

Although the intensity of an OLED with a structured PEDOT layer is in general higher, when the same voltage is applied, it is not caused by diffraction of the grating, as the higher intensity is also observed for the direction parallel to the grating, where no diffraction occurs. The higher intensity could be caused by a higher current density. This is also confirmed by the V-I curve as indicated in Figure 6.5a. Since a higher current is flowing through the structured OLED sample for the same voltage, the emission is also expected to be higher. Several reasons for a higher current can be given, like larger surface contact between the PEDOT and PPV, a lower average layer thickness of the PEDOT layer, a chemical modification of the PEDOT properties or scattering by the roughness of the structured PEDOT. The experiments performed are only intended to prove that OLEDs can be made from structured PEDOT layers and do not give conclusive result on the changes in emission.

An V-I and L-I characteristic have been taken from an OLED with a grating period of 800 nm is compared to a reference sample, as depicted in Figure 6.5a and b, respectively. The intensity measurement has been done a normal angles and for a wavelength of 550 nm. The V-I curve of the structured PEDOT layer shows a lower voltage than the reference sample for the same current. Also the intensity of emission of the structured OLED is higher than the
reference sample for the same current, as indicated in Figure 6.5b. This might be caused by lower absorption of the emitted light as the PEDOT layer of the structured film is thinner. Both effects explain the higher angular intensity of emission at a fixed voltage of the structured OLED compared to the reference sample, as indicated in Figure 6.4b.

### 6.1.3 Conclusions

Gratings have been made in PEDOT with a pulsed laser with intensities ranging from 56 to 91 mJ/cm². Unfortunately, the exposed spot is not very homogenous in intensity, allowing only qualitative measurements. It has been observed, that the more PEDOT is removed with increasing intensity. A proof of principle has been given that OLEDs can be made with structured PEDOT layers with grating periods of 800 and 2000 nm.

The angular distribution of OLEDs with a grating in the PEDOT layer with a period Λ of 800 nm seems to be unaffected by the grating. Both emissions from the reference and structured samples show an approximately constant angular intensity distribution. For some OLEDs a higher intensity is observed at large angles from the normal. This is due to scattering by irregularities in the polymer layers or due to light leakage at the side of the OLED. More experiments need to be performed to determine the origin of the increased light output at higher angles. The intensity-voltage curves show no significant differences between the structured and unstructured films.

### 6.2 Structuring PEDOT-PPV layer

#### 6.2.1 Experiment

Similar to the experiments and measurements described in the previous section, OLEDs were made from structured PEDOT-PPV layers. The PPV was spin-coated on-top of the PEDOT layer with a thickness of about a 100 nm. The double layer of PEDOT-PPV was exposed to a similar holographic pattern from a pulsed Nd:YAG laser of 532 nm with a period of 800 nm and 2000 nm. After the exposure, an OLED was constructed and the electrical and optical properties were characterized with a DMS 703.
6.2.2 Results and discussion

Exposure doses ranging from 40 to 112 mJ/cm\(^2\) resulted in diffractive gratings. Energy doses lower than 40 mJ/cm\(^2\) did not show any difference with reference samples. A higher doses than 112 mJ/cm\(^2\) resulted in the removal of the PEDOT-PPV layer. As shown in Figure 6.6, the grating is formed by the deformation of the PPV-layer and the structuring process of the PEDOT layer. The PPV-layer, detaches from the PEDOT-layer forming an array of tunnels. In the SEM-images the degradation of the PEDOT is visible at the center of every tunnel.

Instead the surface of the PPV layer to be effected in a similar way as the PEDOT layer, it forms a tunnel-like structure for every period. One possible reason for the formation of these tunnels could be that the PEDOT degrades before the PPV does. During degradation of the PEDOT, gaseous compounds are created, pushing the PPV layer in this shape \[78, 79\]. Another possibility is mentioned by K. Evans et al. \[80\], which contribute the bending (blister formation) to thermal stresses. The mechanisms involved are elastic buckling due to high radial compressive stresses and interface failure due to axial tensile stresses generated by high axial thermal gradients. Although the PPV-layer detaches partially from the PEDOT layer, functional OLEDs have been made and their emissive characteristics have been measured.

The angular distribution of OLEDs with grating periods of 800 nm and 2000 nm have been measured and compared to reference samples, as shown in Figure 6.7a and b, respectively. The angular intensity distribution for OLEDs with a grating period of 800 nm shows a discontinuity at the normal. This is caused by misalignment in orientation and focus of the microscope of the DMS 703 with the sample. The intensity distribution differs for the direction parallel and perpendicular along the grating compared to the reference OLED. The intensity increases with increasing angle from the normal for both directions. This is most likely due to random scattering, as it is observed for the parallel and perpendicular direction to the grating. For OLEDs with a grating period of 2000 nm, a small oscillation of the intensity as a function of angle is observed, as can be seen in Figure 6.7b. The angular intensity measurement perpendicular to the grating (in the grating direction \(x\)) shows 5 peaks, corresponding to the diffraction orders of a grating with a period of 2000 nm.

An V-I and L-I characteristic have been taken from the OLEDs with a grating of 800 nm.
Figure 6.7: Angular light distribution from an OLED with a structured PEDOT-PPV layer (exposed to 91 mJ/cm$^2$) with a periodicity of 0.8 μm operated at 8V.

![Angular light distribution](image1)

Figure 6.8: Voltage and illuminance dependency as a function of current at normal angle of an OLED with structured PEDOT-PPV by a 91 mJ/cm$^2$ pulse.

![Voltage and illuminance dependency](image2)

and is compared to the reference samples. As can be seen in Figure 6.8a and b respectively, the OLEDs with a grating period of 800 nm give the same voltage for a certain applied current as the reference samples.

### 6.2.3 Conclusions

Gratings have been made in the PEDOT-PPV layer with similar energy levels as for the PEDOT layer. Functional OLEDs have been made from these gratings. The grooves as observed for the PEDOT layer did not appear, however the PPV layer formed a tunnel-like periodic structure.

Similar to the previous section, the angular intensity distribution of the OLEDs have been measured and show irregularities due to scattering or light leakage at the edge of the OLEDs. Large deviations where observed between the samples and no conclusive effects in angular distribution could be determined.

For the OLEDs with grating periods of 2000 nm a small angular intensity modulation is
Micro-structuring of functional layers in OLEDs

6.3 Structuring ITO

6.3.1 experiment

Indium tin oxide (ITO) is a mixture of indium(III) oxide (In$_2$O$_3$) and tin(IV) oxide (SnO$_2$), typically 90% In$_2$O$_3$, 10% SnO$_2$ by weight. It is transparent and colorless in thin layers. A thin layer (200 nm) was sputtered onto the glass substrate.

To structure the ITO a wet holo-lithographic process was used. First, a photo-resist AZ1518 layer of about 1 $\mu$m thickness was spun onto the ITO and the solvent was removed by heating. Next, the photo-resist was exposed to an (holographic) interference pattern from an Argon ion laser with a wavelength of 351 nm with an exposure doses of (70 mW/cm$^2$). Two types of interference patterns were used: a two-beam setup and a three-beam setup, giving a line pattern and hexagonal dots pattern (as shown in Section 2.2.2), respectively. After the holographic exposure, the photo-resist was developed for about 1 minute by a developer solution, which is commercially supplied with the photo-resist and was 50wt% diluted with water. In this case, a positive photo-resists was used, meaning the exposed area gets dissolved. Next, the substrate was washed by isopropanol and water until the sample was cleared from residual developer and dissolved resist. Due to the pattern in the photo-resist, the ITO-layer is partially uncovered, making it accessible for etching fluids. Amorphous ITO was etched with a 6.67 g/l oxalic acid for 7 minutes at room temperature. Crystalline ITO was etched for 1 minute at 40$^\circ$C with a 6 M HCl and 6wt% FeCl$_3$ solution. Finally, the residual photo-resist was removed by washing the substrate with acetone.

6.3.2 Results

After patterning and developing the photo-resist, it obtains the similar pattern to the exposed interference pattern in pitch and structure, as indicated in Figure 6.9. Depending on the interference pattern either a line pattern (Figure 6.9a) or a hexagonal pattern (Figure 6.9b) is obtained.

Figure 6.9: Patterned and developed photo-resist structure with (a) a line pattern and (b) a hexagonal pattern.
Several holographic structures were made in the ITO-layers with different crystallinity. The higher the crystallinity, the slower the etching-process and the rougher the ITO-structure is. In Figure 6.10b the ITO-crystals are clearly visible, while the amorphous ITO (Figure 6.10a) does not show any crystal grains.

6.3.3 Conclusions

Gratings have been made in the ITO-layer by a wet etching process. After exposing a photo-resist with a three-beam, hexagonal or a two-beam, line interference pattern, the negative polymer structure was obtained. Etching the ITO and removing the photo-resist resulted in patterned ITO substrates.

6.4 Outlook

Functional OLEDs can be made from structured PEDOT and PPV layers. Potentially, these gratings can result in more efficient OLEDs and OLEDs with a controlled angular emission. However, only qualitative experiments have been carried out so far. To draw conclusions on the improvement of the out-coupling, quantitative experiments need to be performed. Therefore, some critical barriers need to be overcome first, mainly concerning the reproducibility.

To improve reproducibility, the exposure spot of the pulsed laser needs to be more homogenous in intensity. The spot-size of the laser was focussed to only a few millimeters in size to achieve high intensities. The measured spot-size was 1 mm in diameter at normal angle, which is too close to the spot-size.

Overall, a relative high voltage needs to be applied the OLEDs (above 6V). This poor performance is due to time delay between coating the layers, the holographic exposure and the encapsulation, which resulted in degradation (oxidation) of the PPV. Ideally, the holographic exposure should be done in an oxygen and water free atmosphere. Still, since the reference OLEDs have undergone the same processing conditions (and oxidation), a comparison can be made with the structured OLEDs.

The detachment of the PPV-layer from the PEDOT-layer has a very negative influence on the conductance and consequentially the intensity of emission, which is undesirable for
lighting applications. However, applications for micro-channels in conductive and emissive polymers can be envisioned in the field of biosensors and analytical measurements.

No OLEDs have been made from the structured ITO substrates. However, McGehee et al. [75] report that by putting a Bragg-grating in the ITO-layer to scatter the waveguided light out an efficiency gain with a factor of 1.25 is obtained. Instead of using reactive-ion etching, a more simple wet etching process is used. Many grating parameters can be investigated to optimize the efficiency of OLEDs, like the grating period and layer thickness. Removing ITO by etching reduces the conductivity and careful considerations need to be taken to avoid the reduction of efficiency. As shown, the amorphous ITO is much easier and faster to etch and gives smooth ITO patterns. However, for OLEDs crystalline ITO is commonly used, since it has a higher conductivity.

It has been shown that the function layers of an OLED can be structured by holography. Different grating periods and patterns have been created and functional OLEDs have been made. Future research should be done to investigate the effects of these gratings on the light distribution and efficiency. Under the influence of a grating, not only the direction of the emitted light changes, but also the conductivity and quantum efficiency. Applying a grating inside an OLEDs will require the optimization of the layer thicknesses of the functional layer and the grating parameters.
References


REFERENCES


Appendix A

A.1 Elliptical polarization holograms

In general, the interference pattern of two orthogonal elliptical polarized states with ellipticity angle $\chi$ and orientation angle $\psi$ results in a modulation between four sequential polarization states, as shown in the picture A.1. The interference pattern of plane waves, with a polarization state described by $\tan \chi$ and $\psi$, and its orthogonal polarization state $-\tan \chi$ and $\psi - \pi/2$, results in an interference pattern with the following polarization states.

1. A linear polarization state with orientation angle $\psi + \pi/4$.
2. An elliptical polarization state with ellipticity $\tan \chi = -1 + \frac{b}{a}$ and orientation angle $\psi$.
3. A linear polarization state with orientation angle $\psi - \pi/4$.
4. An elliptical polarization state with ellipticity $\tan \chi = 1 - \frac{b}{a}$ and orientation angle $\psi - \pi/2$.

The orientation of the interference patterns rotate continuously along the grating, unless the polarization states of the orthogonal recording plane waves are linear. If purely linear polarized light is used for recording, the orientation angle alternates between $\psi$ and $\psi + \pi/2$ at the circular polarization states ($S_3 = -1$ or $S_3 = 1$). The ellipticity in general modulates from zero to the ellipticity of $1 - b/a$ of the recording beams.

A.2 Derivation of the transfer matrix of polarization gratings

R.M.A. Azzam et al.[47] describe the evolution of the ellipse of polarization in anisotropic media. Using a basis of eigenpolarizations, the equation of propagation for the Jones vector is expanded into two coupled first-order ordinary differential equations A.1.

\[
\begin{align*}
\frac{dE_u}{dz} &= n_{1,1}E_u + n_{1,2}E_v \\
\frac{dE_v}{dz} &= n_{2,1}E_u + n_{2,2}E_v
\end{align*}
\] (A.1)

By assuming that the refractive index does not change over the thickness of the layer (the propagation direction) the differential equations are solved in terms of eigenpolarizations, eigenvalues $\xi_{e1}$ and $\xi_{e2}$ and birefringence $\Delta n$, resulting in the N-matrix A.2.
Appendix A

Figure A.1: The interference pattern of two orthogonal elliptical polarization states (shown on top) in four steps (shown in the bottom).

\[
\mathbf{n} = \begin{bmatrix}
-i \frac{2\pi}{\lambda} \left( n_0 + \frac{\xi_{e1} + \xi_{e2}}{\xi_{e1} - \xi_{e2}} \Delta n \right) \\
-i \frac{4\pi}{\lambda} \frac{\Delta n}{\xi_{e1} - \xi_{e2}} \\
-i \frac{2\pi}{\lambda} \left( n_0 - \frac{\xi_{e1} + \xi_{e2}}{\xi_{e1} - \xi_{e2}} \Delta n \right)
\end{bmatrix}
\] (A.2)

with:

\[
\xi_{e1} = \frac{\tan \psi + i \tan \chi}{1 - i \tan \psi \tan \chi}, \quad \xi_{e2} = \frac{-1 - i \tan \psi \tan \chi}{\tan \psi - i \tan \chi}
\] (A.3)

Substitution of the eigenpolarizations results in the following \(\mathbf{N}\)-matrix:

\[
\mathbf{n} = -i \frac{2\pi}{\lambda} \begin{bmatrix}
n_0 - \Delta n \cos(2\chi) \cos(2\psi) \\
\frac{\Delta n}{2} (\sin(2\psi) \cos(2\chi) - i \sin(2\chi)) \\
n_0 - \Delta n \cos(2\chi) \cos(2\psi)
\end{bmatrix}
\] (A.4)

Substituting the Stokes parameter for the orientation and ellipticity angles (Equation 1.6) and using \(\mathbf{T} = \exp(\mathbf{n}d)\) results in transfer matrix 5.12. To obtain Equation 5.12 the birefringence has to be split into a linear and circular part. The justification for this split is discussed in Section 5.3.

A.3 Polarization-selective diffraction in Stokes parameters

As mentioned in Chapter 5, the diffraction efficiencies \(\eta_m\) of every order \(m\) are obtained by the far-field \(\mathbf{D}_m\) by:

\[
\eta_m = |\mathbf{D}_m|^2
\] (A.5)

This far-field is obtained by applying the Fourier transform of the transfer matrix for every diffraction order \(m\). Assuming that the incoming electric field \(\mathbf{E}_\text{in}\) is constant in the grating direction \(x\) every element of \(\mathbf{T}(x)\) can be integrated separately. In this case the general solution of the far-field diffraction can be written as:
\[ \mathbf{D}_m = \left( \frac{T'_{1,1}(m)E_x + T'_{1,2}(m)E_y}{T'_{2,1}(m)E_x + T'_{2,2}(m)E_y} \right) \] (A.6)

With:

\[ T'_m = \frac{1}{\Lambda} \int_0^\Lambda T(x) \exp \left( \frac{-i2\pi mx}{\Lambda} \right) \, dx \] (A.7)

The diffraction efficiencies are therefore:

\[ \eta_m = |\mathbf{D}_m|^2 \] (A.8)

\[ \eta_m = (T'_{1,1}(m)E_x + T'_{1,2}(m)E_y) \left( \frac{T'_{1,1}(m)E_x + T'_{1,2}(m)E_y}{T'_{2,1}(m)E_x + T'_{2,2}(m)E_y} \right) \]

\[ \eta_m = \left( |T'_{1,1}(m)|^2 + |T'_{2,1}(m)|^2 \right) |E_x|^2 + \left( |T'_{1,2}(m)|^2 + |T'_{2,2}(m)|^2 \right) |E_y|^2 + \]

\[ (T'_{1,1}(m)T'_{1,2}(m) + T'_{2,1}(m)T'_{2,2}(m)) E_x E_y + \]

\[ (T'_{1,1}(m)T'_{1,2}(m) + T'_{2,1}(m)T'_{2,2}(m)) E'_x E'_y \]

The Stokes parameters can be substituted for the electric field components, resulting in:

\[ \eta_m = 0.5 \left( |T'_{1,1}(m)|^2 + |T'_{2,1}(m)|^2 \right) \left( S_0 + S_1 \right) + \]

\[ 0.5 \left( |T'_{2,1}(m)|^2 + |T'_{2,2}(m)|^2 \right) \left( S_0 - S_1 \right) + \]

\[ 0.5 \left( T'_{1,1}(m)T'_{1,2}(m) + T'_{2,1}(m)T'_{2,2}(m) \right) \left( S_2 - iS_3 \right) + \]

\[ 0.5 \left( T'_{1,1}(m)T'_{1,2}(m) + T'_{2,1}(m)T'_{2,2}(m) \right) \left( S_2 + iS_3 \right) \]

Rewriting leads to:

\[ \eta_m = 0.5 \left( |T'_{1,1}(m)|^2 + |T'_{2,1}(m)|^2 + |T'_{2,2}(m)|^2 \right) \left( S_0 + \right) \]

\[ 0.5 \left( |T'_{1,1}(m)|^2 - |T'_{2,1}(m)|^2 + |T'_{2,2}(m)|^2 \right) \left( S_1 + \right) \]

\[ 0.5 \left( T'_{1,1}(m)T'_{1,2}(m) + T'_{2,1}(m)T'_{2,2}(m) + T'_{1,1}(m)T'_{1,2}(m) + T'_{2,1}(m)T'_{2,2}(m) \right) \left( S_2 + \right) \]

\[ -i0.5 \left( T'_{1,1}(m)T'_{1,2}(m) + T'_{2,1}(m)T'_{2,2}(m) - T'_{1,1}(m)T'_{1,2}(m) - T'_{2,1}(m)T'_{2,2}(m) \right) \left( S_3 \right) \]

To normalize the diffraction efficiencies, they are divided by the intensity \( S_0 \), obtaining Equation 5.17 of Chapter 5.

### A.4 Third order Fourier approximation of the transfer matrix

To analytically integrate the transfer matrix with respect to \( x \) in Equation 5.28, a third order Fourier approximation is used for the \( \cos(F) \) and the \( \sin(F)/F \) terms.

\[ \cos(F) \approx \frac{1}{2} \left( \cos(N_{lin}) + \cos(N_{cir}) + (\cos(N_{lin}) - \cos(N_{cir})) \cos \left( \frac{4\pi x}{\Lambda} \right) \right) \] (A.10)

\[ \frac{\sin(F)}{F} \approx \frac{1}{2} \left( \frac{\sin(N_{cir})}{N_{cir}} + \frac{\sin(N_{lin})}{N_{lin}} + \frac{\sin(N_{cir})}{N_{cir}} - \frac{\sin(N_{lin})}{N_{lin}} \cos \left( \frac{4\pi x}{\Lambda} \right) \right) \] (A.11)

These approximations are substituted in transfer matrix in Equation 5.28 allowing them to be analytically integrated.
Samenvatting

Polarisatoren worden veel gebruikt om gepolariseerd licht te maken in apparaten zoals vloeibaar kristallijnen beeldschermen. Deze beeldschermen zijn afhankelijk van gepolariseerd licht om het aan en uit te kunnen schakelen. Lineair gepolariseerd licht wordt normaliter gemaakt door lineair gepolariseerd licht in de ongewenste richting te absorberen. Hierdoor gaat ongeveer 50% van het licht verloren. Nog eens 66% gaat verloren door het gebruik van kleurenfilters om de primaire kleuren rood, groen en blauw te maken. Beide absorberende optische elementen dragen bij aan het feit dat een dergelijk beeldscherm erg inefficiënt is in licht-gebruik. Typisch bereikt slechts 5% van het geproduceerde licht in een display de kijker. In het bijzonder voor mobiele display toepassingen met zo’n vloeibaar kristallijnen beeldscherm, zoals de meeste laptops en mobiele telefoons, kan deze inefficiëntie het gebruik bij een gegeven capaciteit van een batterij significant verkorten.

In dit proefschrift wordt een alternatieve polarisator gepresenteerd, die niet alleen niet-absorberend en dus efficiënter is, maar tevens kleuren scheidt. Er wordt aangetoond dat holografische tralies met een hellende hoek in staat zijn om licht polarisatie selectief uit een golfgeleider te koppelen zonder absorptie. Lineair gepolariseerd licht wordt in de kijkrichting uitgekoppeld, terwijl het andere gedeelte van het licht opgesloten blijft in de golfgeleider. Aangezien de diffractiehoek altijd afhankelijk is van de golflengte (kleur), worden de kleuren automatisch gescheiden en kunnen ze naar het juiste kleurpunt van het beeldscherm gericht worden. Op deze manier kan uiteindelijk een efficiënter beeldscherm gemaakt worden.

Twee verschillende soorten holografische tralies zullen experimenteel onderzocht worden, die beide in staat zijn om polarisatie-selectief en gecollimeerd licht te diffracteren zonder absorptie. Ten eerste wordt een nieuw nano-poreus materiaal gepresenteerd, dat een grote periodieke verandering heeft in brekingsindex en bovendien relatief gemakkelijk om te maken is. De polarisatie selectieve eigenschappen van diffractie worden theoretisch voorspeld door gebruik te maken van de gekoppelde golf-theorie van Kogelnik en worden vervolgens experimenteel geverifieerd. Polarisatie contrasten van 7.5 voor rood, 35 voor groen en 11.2 voor blauw zijn bereikt. Ten tweede worden tralies gepresenteerd die bestaan uit vloeibare kristallen, die gemengd zijn in een polymeren netwerk. Deze tralies zijn in staat polarisatie selectief licht te diffracteren door gebruik te maken van uitgelijnde vloeibare kristallen, waarvan één van de brekingindices overeenkomt met de brekingsindex van het polymeren netwerk. De eerste bevindingen leverden polarisatie contrasten op van ongeveer 10 over het hele golflengte gebied van het zichtbare licht. Bovendien koppelde het licht zich voornamelijk uit in één richting (18 maal meer intens naar voren dan naar achteren) en gecollimeerd tot ongeveer 3°.

Het brekingsindex-profiel van nano-poreuze tralies is nagebootst met een rigoureuze gekoppelde golf-theorie en in overeenstemming gebracht met de diffractie efficiëntie. De hoek-afhankelijke diffractie efficiëntie is gebruikt om de invloed van verschillende parameters te onderzoeken die het brekingsindex-profiel beschrijven. Een blokprofiel met een bepaalde vul-
factor blijkt een geschikte manier te zijn om nano-poroueze tralies met een bepaalde laagdikte na te bootsen.

Een polarisatie selectieve diffractie kan ook bereikt worden door gebruik te maken van zogenaamde polarisatie hologrammen. In dit proefschrift wordt een theoretisch model gepresenteerd, dat de formatie en diffractie van dit type hologrammen beschrijft. Terwijl de meeste theoretische modellen alleen de diffractie en/of formatie van lineair dubbel brekende polarisatie hologrammen beschrijven, worden in dit geval meer algemene polarisatie hologrammen beschreven door middel van modulerende Stokes parameters gecombineerd met lineaire en circulaire dubbel-breking. Met dit theoretische model kunnen polarisatie hologrammen ontworpen worden, die elke vorm van gepolariseerd licht met een hoge efficiëntie kunnen diffracteren. Het ontwerp wordt gemaakt op basis van een interferentiepatroon in combinatie met reactieve materialen die gevoelig zijn voor gepolariseerd licht. Om de toepasbaarheid van het theoretische model aan te tonen, worden er een aantal voorbeelden gepresenteerd, die de polarisatie selectiviteit van polarisatie hologrammen laten zien. Uiteindelijk worden de polarisatie hologrammen geclasseerd op basis van het interferentie patroon en de vorming van een patroon in dubbel-breking index.

Het uitkoppelen van licht d.m.v. tralies om de efficiëntie van beeldschermen te verbeteren is niet beperkt tot vloeibaar kristallijne beeldschermen. Deze tralies kunnen ook gebruikt worden om de efficiëntie van beeldschermen te verbeteren die gebruik maken van organische lichtgevende diodes (OLED’s). Hoewel de polarisatie-selectiviteit van minder belang is voor dit type beeldschermen, kunnen tralies de efficiëntie verhogen door licht uit te koppelen, dat gevangen is in de OLED vanwege totale interne reflectie. Er wordt aangetoond dat het mogelijk is om tralies te maken in de organische lagen van de OLED. Bovendien zijn er tralies gemaakt in de geleidende indium-tin-oxide laag. Verschillende soorten patronen zijn aangebracht in verschillende soorten indium-tin-oxide.

Samenvattend wordt gedemonstreerd dat nauwkeurig ontworpen tralies gebruikt kunnen worden om de efficiëntie van vloeibaar kristallijnen en OLED beeldschermen te verbeteren. Er is een goede overeenkomst tussen de theoretische voorspellingen en de praktische resultaten van de nano-poroueze en vloeibaar kristallijnen tralies met een hellende hoek voor polarisatie-selectieve uitkoppeling van golfgeleidend licht. Polarisatie hologrammen zijn zeer geschikt voor het efficiënt breken van licht met een vrij te bepalen polarisatie toestand.
Technology Assessment

Recently, telecommunications have made huge steps in terms of data transfer rates leading to more and better communication means. Furthermore, there are no signs yet which indicate that this revolution has already ended. Mobile phones, personal digital agendas (PDAs) and cameras are good examples of recent devices, that facilitate the need to communicate easily and frequently.

Portability, which is key for these devices, inherently implies the requirement of a low power consumption, as the capacity of portable batteries is limited. Yet a colorful display with a high contrast and brightness is demanded for such devices to optimally convey large amounts of information. As a result, a significant part of the total power consumption is contributed to the display.

The high power consumption of current displays is caused by their low efficiency. The performance of optical devices, such as displays, depends critically on controlling the direction, polarization and collimation of light over the entire range of visible wavelengths. Typically, the absorbing polarizers and color filters, that are currently used in a display make these devices optically inefficient. A need of new non-absorbing optical elements therefore exists.

Polarization-selective gratings can be used as an alternative to absorbing polarizers in LCDs and to couple out light from a waveguide. The resulting improvement in light management is twofold. First, the light that is not coupled out from a waveguide, can be recycled inside the waveguide. Second, colors are separated, since the diffraction is wavelength dependent, and can therefore be directed to their corresponding sub-pixels. This avoids absorption of light by color filters.

Holography is a well known technique to make optical diffractive films. Using the interference of a laser, light patterns in intensity or polarization can be written in polymeric materials. However, for large scale productions the preference is given to use masks, as they are more homogeneous in intensity. Lasers usually have a Gaussian intensity distribution through the cross-section of the beam. In principle, an equivalent phase mask can be produced for every holographic interfere pattern. The reason to use holography during this PhD-project is that it has superior flexibility in producing a large variety of interference patterns and meets the lab scale requirements.

The development of liquid crystal polarization gratings opens up a whole new field of efficient displays. The unique characteristics of this type of grating with respect to diffraction efficiency and polarization-selectivity in combination with their ability to switch, enables the production of e.g. efficient low-power projection displays. Current projection displays, like most displays, use absorbing polarizers and color filters. Besides the inherent low efficiency, this absorption is especially disadvantageous for projection displays, because of heat that is produced by absorption. Active heat management is typically required for these devices. Wavelength dependent diffraction, polarization-selectivity, and a high diffraction efficiency
are properties of a polarization grating, which make the use of absorbing polarizers and color filters redundant for projection displays. This increases the efficiency to the extent that low power light sources, like LEDs, suffice. In turn, this makes the production of portable projection displays practical.

Although theoretical predictions reveal the possibility to tailor the polarization-selective diffraction of any state of polarization, a lot of research needs to be performed on polarization-sensitive materials. Linear birefringent materials have seen over three decades of intensive development, yielding a widespread commercial availability. Suitable circular birefringent materials, on the other hand, are not readily available. Although many biological materials are circularly birefringent (also referred to as optical-active or chiral materials), their magnitude of birefringence is usually an order of magnitude lower compared to the linear birefringent liquid crystals. Furthermore, left- and right-handed versions should be available. For these reasons, a lot of development is still required on the material properties of polarization gratings.

In the near future, OLEDs might serve as an alternative to the current backlight systems of LCDs. Currently, OLEDs have a poor light management since a lot of generated light remains trapped inside an OLED due to total internal reflections. Applying a grating inside its functional layers might improve the efficiency and control the angular emission of an OLED. In this thesis a proof of principle is presented of creating gratings in the functional layers of an OLED.
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Curriculum Vitae

Chris van Heesch was born in Eindhoven February 2, 1979. After finishing secondary school at Zwijsen College (Veghel), in 1997 he started studying Chemistry and Chemical Engineering at Eindhoven University of Technology. With a specialization in polymer chemistry and technology he did his Master’s thesis on broadening the reflection band of cholesteric color filters at Philips Research in Eindhoven. Afterwards he worked for a three months traineeship at Unilever Research in Vlaardingen on an on-line detection method of oil exudation for a high throughput system.

In March 2003 Chris returned to Eindhoven University of Technology for his PhD project at the group of Polymer for Information and Communication Technology of Prof.Dr. Dick Broer and Dr.Ing. Cees Bastiaansen. During his PhD he went for three months to North Carolina working together with Prof.Dr. Micheal Escuti at the NC State University mainly on the theory behind the diffraction of polarization holograms. Furthermore, a collaboration was setup with the University of Zaragoza on outcoupling structures for OLEDs, leading to the PhD-study reported in this thesis.