Complex Oxide Photonic Crystals

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Licentiate Thesis
Stockholm, Sweden 2009
Akademisk avhandling som med tillstånd av Kungl Tekniska högskolan framlägges till offentlig granskning för avläggande av teknologie licentiatexamen torsdagen den 21 september 2009 klockan 10.15 i C4, Electrum 229, Kungl Tekniska högskolan, Stockholm.

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Tryck: Kista Snabbtryck AB
Abstract

Microphotonics has been offering a body of ideas to prospective applications in optics. Among those, the concept of photonic integrated circuits (PIC’s) has recently spurred a substantial excitement into the scientific community. Realisation of the PIC’s becomes feasible as the size shrinkage of the optical elements is accomplished. The elements based on photonic crystals (PCs) represent promising candidacy for manufacture of PIC’s.

This thesis is devoted to tailoring of optical properties and advanced modelling of two types of photonic crystals: \((\text{Bi}_3\text{Fe}_5\text{O}_{12}/\text{Sm}_3\text{Ga}_5\text{O}_{12})^5\) and \((\text{TiO}_2/\text{Er}_2\text{O}_3)^6\) potentially applicable in the role optical isolators and optical amplifiers, respectively. Deposition conditions of titanium dioxide were first investigated to maximise refractive index and minimise absorption as well as surface roughness of titania films. It was done employing three routines: deposition at elevated substrate temperatures, regular annealing in thermodynamically equilibrium conditions and rapid thermal annealing (RTA). RTA at 500°C was shown to provide the best optical performance giving a refractive index of 2.53, an absorption coefficient of 404 cm\(^{-1}\) and a root-mean-square surface roughness of 0.6 nm.

Advanced modelling of transmittance and Faraday rotation for the PCs \((\text{Bi}_3\text{Fe}_5\text{O}_{12}/\text{Sm}_3\text{Ga}_5\text{O}_{12})^5\) and \((\text{TiO}_2/\text{Er}_2\text{O}_3)^6\) was done using the 4 × 4 matrix formalism of Višňovský. The simulations for the constituent materials in the forms of single films were performed using the Swanepoel and Višňovský formulae. This enabled generation of the dispersion relations for diagonal and off-diagonal elements of the permittivity tensors relating to the materials. These dispersion relations were utilised to produce dispersion relations for complex refractive indices of the materials. Integration of the complex refractive indices into the 4 × 4 matrix formalism allowed computation of transmittance and Faraday rotation of the PCs. The simulation results were found to be in a good agreement with the experimental ones proving such a simulation approach is an excellent means of engineering PCs.
I would like to express my sincere gratitude to the following persons:

Prof. Alex Grishin, my supervisor, for his wise supervision over me, help in understanding the theory, giant efforts to make the chemical mind transform to a physical one, and for being the great mentor in my life-classes.

Dr. Sergei Popov, my co-supervisor, for his kind help, opening a door to my knowledge of optics, his crucial suggestions on everyday life and for being a good man in all respects.

Dr. Sergiy Khartsev, our senior researcher, for the best experimental suggestions, valuable advices and invaluable help.

My family, for their love, eternal and inspiring support at every step of my life.

Kamilia, for my happiness.
Notations

\( \text{a, b, c} \) fundamental translation vectors
\( B \) magnetic induction
\( c \) speed of light
\( d \) film thickness
\( E \) electric field
\( E_0 \) electric field amplitude
\( e \) integer for oscillation maxima or half-integer for oscillation minima
\( f \) oscillator strength
\( f_{\pm} \) oscillator strength for positive and negative transitions
\( H, K, L \) fundamental translation vectors of the reciprocal lattice
\( I, \Im \) light intensity
\( J \) Jones vector
\( J_R \) Jones vector for right-handed circularly polarised light
\( J_L \) Jones vector for left-handed circularly polarised light
\( k \) extinction coefficient \( (k = -3m\sqrt{\varepsilon_{xx}}) \)
\( M \) transfer matrix of the whole multilayer
\( N \) complex refractive index \( (N = n - ik) \)
\( N_{\pm} \) complex refractive indices for left- and right-handed circularly polarized light
\( n \) refractive index \( (n = \Re\sqrt{\varepsilon_{xx}}) \)
\( r \) position vector
\( s \) refractive index of substrate
\( T \) transmittance
\( T_{(i-1,i)} \) transfer matrix from layer \((i-1)\) to layer \((i)\)
\( T_s \) substrate temperature
\( t \) time
\( \hat{x}, \hat{y}, \hat{z} \) Cartesian unit vectors
\( x \) optical path difference
\( \Gamma \) full width at half maximum of resonance
\( 2\Delta \) splitting of excited state
\( \theta \) angle between the semi-major axis of ellipse and \( x \)-axis
\( \Theta_F \) total angle of Faraday rotation
\( \alpha \) absorption coefficient
\( \beta \) propagation constant \( (\beta = \omega Nd/c) \)
\( \gamma \) wave vector
\( \hat{\xi} \) tensor of relative permittivity
\( \theta_F \) angle of Faraday rotation per film thickness
\( \lambda \) wavelength of light
\( \xi \) angle between the vector of electric field and \( x \)-axis
\( \sigma \) root-mean-square surface roughness
\( \phi \) phase difference
\[ \phi_x, \phi_y \] phases of waves with their electric field parallel to \( x \)- and \( y \)-axes

\[ \psi \] angle of ellipticity

\[ \omega \] angular frequency

\[ \omega_0 \] resonance angular frequency

\[ \omega_{0 \pm} \] resonance angular frequency for left- and right-handed circularly polarised light \((\omega_{0 \pm} = \omega_0 \pm \Delta)\)

\[ \omega_p \] plasma frequency
### Abbreviations

<table>
<thead>
<tr>
<th>Abbreviation</th>
<th>Description</th>
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<tbody>
<tr>
<td>AFM</td>
<td>atomic force microscope</td>
</tr>
<tr>
<td>BIG</td>
<td>bismuth iron garnet (Bi₃Fe₅O₁₂)</td>
</tr>
<tr>
<td>CG</td>
<td>Corning glass</td>
</tr>
<tr>
<td>EDFA</td>
<td>erbium-doped fibre amplifier</td>
</tr>
<tr>
<td>FR</td>
<td>Faraday rotation</td>
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<tr>
<td>FTIR</td>
<td>Fourier transform infrared</td>
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<tr>
<td>GGG</td>
<td>gadolinium gallium garnet (Gd₃Ga₅O₁₂)</td>
</tr>
<tr>
<td>GMZGGG</td>
<td>calcium-, magnesium-, zirconium-doped gadolinium gallium garnet (Ca,Mg,Zr:Gd₃Ga₅O₁₂)</td>
</tr>
<tr>
<td>IG</td>
<td>iron garnet</td>
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<tr>
<td>IR</td>
<td>infrared</td>
</tr>
<tr>
<td>LCP</td>
<td>left-handed circularly polarized</td>
</tr>
<tr>
<td>MO</td>
<td>magneto-optical</td>
</tr>
<tr>
<td>MOPC</td>
<td>magneto-optical photonic crystal</td>
</tr>
<tr>
<td>PC</td>
<td>photonic crystal</td>
</tr>
<tr>
<td>PIC</td>
<td>photonic integrated circuit</td>
</tr>
<tr>
<td>PLD</td>
<td>pulsed laser deposition</td>
</tr>
<tr>
<td>RCP</td>
<td>right-handed circularly polarized</td>
</tr>
<tr>
<td>RE</td>
<td>rare earth</td>
</tr>
<tr>
<td>REIG</td>
<td>rare earth iron garnet (RE₃Fe₅O₁₂)</td>
</tr>
<tr>
<td>REIG:Al/Ga/Sc</td>
<td>rare earth iron garnet substituted by Al, Ga or Sc</td>
</tr>
<tr>
<td>RF</td>
<td>radio-frequency</td>
</tr>
<tr>
<td>RMS</td>
<td>root-mean-square</td>
</tr>
<tr>
<td>RTA</td>
<td>rapid thermal annealing</td>
</tr>
<tr>
<td>SGG</td>
<td>samarium gallium garnet (Sm₃Ga₅O₁₂)</td>
</tr>
<tr>
<td>XRD</td>
<td>X-ray diffraction</td>
</tr>
<tr>
<td>YIG</td>
<td>yttrium iron garnet (Y₃Fe₅O₁₂)</td>
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Part I

Preliminaries
Chapter 1

Introduction

1.1 Background

The era of silicon electronics is gradually coming to an end. The intrinsic limits of silicon-based devices in terms of bandwidth and operational speed shall preclude the currently-used devices from further development.

On the contrary, optical media allow much more benefits in comparison with silicon-based analogues. The demand for a size shrinkage of elements in optical systems, however, represents a formidable challenge. It is the driving force of a transition from discrete to integrated optical components. That is where microphotonics comes in bringing out the concept of photonic integrated circuit (PIC). The possibility of having active and passive optical components on a single chip is by far the most desired.

Since their discovery [33, 68], photonic crystals (PCs) has gained considerable attention owing to their ability to control spontaneous emission, bend light, trap photons, etc. Such properties enable PC-based elements to occupy the niche of attractive candidates for monolithic integration into PICs. There are two components of PICs being of prime importance: optical amplifiers and optical isolators. In the core of the former lies Er$_2$O$_3$ which acts as a light emitting medium, whereas the latter are chiefly built of magneto-optical (MO) materials such as iron garnets (IGs).

1.2 Aims and Objectives

Erbium-doped fibre amplifiers (EDFAs) represent well-established products on the market of optical telecommunication systems. Most frequently, such amplifiers exploit Er$_2$O$_3$-doped media since they provide high gain for lasing in the C- and L-bands. Despite such a celebrated applicability, EDFAs possess one intrinsic drawback, namely high costs.

One way to dispose of the high costs is to use Er$_2$O$_3$ thin films which showed a
Chapter 1. Introduction

Figure 1.1. Schematic of optical isolator. Black arrows denote electric field vectors of the beams, blue lines are the transmission axes of polariser (transmission axis positioned vertically) and analyser (transmission axis positioned at 45°).

substantial potential for use in high-gain waveguide amplifiers \[28, 62\]. Inclusion of Er$_2$O$_3$ as a cavity layer in a PC will compel the light to localise inside the cavity. This will thereby yield higher intensity of luminescence in comparison with a single film of Er$_2$O$_3$ \[25\].

The Faraday rotation (FR) is the very effect used in optical isolators to preclude damage of laser. The operational principle of an optical isolator is illustrated in Fig. 1.1. A linearly-polarised laser beam passes through a polariser whose transmission axis is parallel to the electric field (E-field) vector of the beam. Owing to the following advance through a Faraday rotator, the beam gets its E-field vector rotated by 45°. After passing through an analyser, reflection of the beam takes place from the mirror. The beam travels further through the analyser and the Faraday rotator acquiring its E-field vector rotated by 90°. The polarisation state of the beam is now horizontal. On the other hand, transmission axis of the polariser is vertical which does not allow the beam to come through. This results in no damage of the laser.

Optical isolators used presently are rather large. In order to reduce their dimensions, much higher FR is needed. This may be attained using PCs composed of IGs \[27, 36, 38, 39\].

Owing to their small dimensions, the PC-based optical amplifiers and optical isolators are attractive candidates for integration in PICs. However, several problems have to be explored prior to the actual implementation.

Thus, the following thesis is dedicated to a thorough investigation of the two issues. The first one lies in tailoring optical properties of (Er$_2$O$_3$/TiO$_2$) m PCs. This is done by finding conditions of fabrication of TiO$_2$ thin films that provide highest refractive index, lowest absorption and lowest surface roughness of the films. The second issue comprises an advanced modelling of transmittance for the (Er$_2$O$_3$/TiO$_2$)$^6$ PC and (Bi$_3$Fe$_5$O$_{12}$/Sm$_3$Ga$_5$O$_{12}$)$^5$ magneto-optical photonic crystal (MOPC) as well as FR for the (Bi$_3$Fe$_5$O$_{12}$/Sm$_3$Ga$_5$O$_{12}$)$^5$ MOPC \[27\]. Such
modelling may be regarded as a stable approach to engineering the PCs with superior performance.

1.3 Outline

The discourse of the thesis is structured in the following fashion.

In Chapter 2, theoretical preliminaries of polarised light, the Jones calculus and the $4 \times 4$ matrix formalism are summarised in order to derive a means of modelling transmittance and Faraday rotation for the single films as well as photonic crystals.

Material overview is presented in Chapter 3 with a special emphasis on the reason for choice of the materials.

Characterisation techniques used for the material analysis are described in Chapter 4.

Chapter 5 is devoted to the investigation of optical properties of TiO$_2$.

Chapter 6 elaborates on the approach to the simulations of transmittance and Faraday rotation of the single film as well as photonic crystal structures.

Finally, conclusions of the thesis are drawn in Chapter 7.
Chapter 2

Theoretical Background

Throughout this theoretical treatise, we restrict our consideration to normal incidence of light upon homogeneous, linear media with their magnetisation, if any, perpendicular to the surface of a medium ($\gamma \parallel$ magnetisation $\parallel \hat{z} \perp$ to the surface, $\gamma$ is wave vector).

In 1873, James Clerk Maxwell published the set of four differential equations which govern propagation of light \[46\]. We take two of them:

$$\nabla \times \mathbf{E} = -\frac{\partial \mathbf{B}}{\partial t}, \quad (2.1)$$

$$\nabla \times \mathbf{B} = \frac{1}{c^2} \varepsilon \frac{\partial \mathbf{E}}{\partial t}, \quad (2.2)$$

and after some algebraic manipulations arrive at the wave equation:

$$\nabla^2 \mathbf{E} - \nabla (\nabla \cdot \mathbf{E}) = \frac{1}{c^2} \varepsilon \frac{\partial^2 \mathbf{E}}{\partial t^2}, \quad (2.3)$$

where $c$ denotes the speed of light, $t$, time, $\mathbf{B}$ is magnetic induction, $\mathbf{E}$ is electric field, and $\hat{\varepsilon}$ is tensor of relative permittivity.

The tensor of relative permittivity

$$\hat{\varepsilon} = \begin{pmatrix} \varepsilon_{xx} & \varepsilon_{xy} & 0 \\ -\varepsilon_{xy} & \varepsilon_{xx} & 0 \\ 0 & 0 & \varepsilon_{zz} \end{pmatrix} \quad (2.4)$$

defines the response of a medium to electromagnetic disturbances which are the solutions to the wave equation in the form of plane waves

$$\mathbf{E} = \mathbf{E}_0 \exp[i(\gamma \cdot \mathbf{r} - \omega t)], \quad (2.5)$$

$\mathbf{E}_0$ being complex electric field amplitude, $\omega$, angular frequency and $\mathbf{r}$, position vector.
Chapter 2. Theoretical Background

Figure 2.1. Schematic drawing of a diamagnetic transition with the following notations: 
\(|g|\), ground state; |e>, excited state split into two |e+> and |e−> by 2\(\Delta\) — energy of splitting of excited state; \(\omega_0\), resonance frequency; \(\omega_{0\pm}\), resonance frequency of left- and right-handed circularly polarized light.

The expressions for diagonal \(\varepsilon_{xx}\) and off-diagonal \(\varepsilon_{xy}\) elements of Eq. 2.4 can be derived using the density matrix formalism in the frame of electric dipole approximation [6]. If one assumes a diamagnetic character of MO interaction including electron transition from the spin singlet ground state to the excited state split by spin-orbit coupling into two (see Fig. 2.1), they get:

\[
\varepsilon_{xx} = 1 + \omega_p^2 \sum \frac{f_\pm}{\omega_{0\pm}^2} \frac{\omega_{0\pm}^2 - \omega^2 + \Gamma^2 - 2i\omega\Gamma}{(\omega_{0\pm}^2 - \omega^2 + \Gamma^2)^2 + 4\omega^2\Gamma^2} 
\]

and

\[
\varepsilon_{xy} = i\omega_p^2 \sum \frac{(-1)^{\pm} f_\pm}{\omega_{0\pm}^2} \times \frac{\omega(\omega_{0\pm}^2 - \omega^2 - \Gamma^2) - i\Gamma(\omega_{0\pm}^2 + \omega^2 + \Gamma^2)}{(\omega_{0\pm}^2 - \omega^2 + \Gamma^2)^2 + 4\omega^2\Gamma^2},
\]

where \(\omega_p\) is plasma frequency, \(\omega_{0\pm} = \omega_0 \pm \Delta\), the resonance angular frequency for left- and right-handed circularly polarized light, \(\omega_0\), resonance angular frequency, \(\Gamma\), half-linewidth of the transition between the ground state \(|g|\) and the excited state \(|e\rangle\) split by 2\(\Delta\) split energy into the two states \(|e\pm\rangle\), \(f_\pm \approx (f/2)(1 \pm \Delta/\omega_0)\), oscillator strengths for left- and right-handed circular polarisations [4]. In Eqs 2.6 and 2.7 the sum over all of the possible excited states is implied.
Let us recourse to the following assumptions: \( \omega_0 \gg \Delta, |\omega_0^2 - \omega^2| \gg \Gamma^2, |\omega_0^2 - \omega^2| \gg 2\omega \Gamma \) and \( n^2 \gg k^2 \), where \( n = \Re(\sqrt{\varepsilon_{xx}}) \) is refractive index and \( k = -\Im(\sqrt{\varepsilon_{xx}}) \) is extinction coefficient. Assuming also a single dipole transition, Eq. 2.6 can be simplified to give the following relations for \( n \) and \( k \):

\[
\begin{align*}
n^2(\lambda) & \approx 1 + \frac{\Omega^2}{1 - \Lambda^2}, \\
k(\lambda) & \approx \frac{\Lambda \Gamma}{\omega_0} \left( \frac{\Omega}{1 - \Lambda^2} \right)^2 \left( \frac{1 - \Lambda^2}{1 - \Lambda^2 + \Omega^2} \right)^{1/2},
\end{align*}
\]

where \( \Lambda = \lambda_0 / \lambda \) and \( \Omega = \omega_p \sqrt{J} / \omega_0 \). Eq. 2.8 is the well-known Sellmeier equation to be used later.

2.1 States of Polarisation

Any state of polarisation can be synthesised from two orthogonal disturbances representing plane waves with a certain phase difference \( \phi \). Consider two orthogonal disturbances propagating along the \( z \)-axis:

\[
\begin{align*}
E_x &= E_{0x} \cos(\gamma z - \omega t), \\
E_y &= E_{0y} \cos(\gamma z - \omega t + \phi),
\end{align*}
\]

where \( E_{0x} = \hat{x} E_{0x} \) and \( E_{0y} = \hat{y} E_{0y} \) (\( \hat{x} \) and \( \hat{y} \) are the Cartesian unit vectors). Eq. 2.11 can be expanded in terms of cosine of angle sum. Substitution of Eq. 2.10 into Eq. 2.11 with some algebra gives

\[
E_y = E_{0y} \cos \phi - \left[ 1 - \left( \frac{E_x}{E_{0x}} \right)^2 \right]^{1/2} \sin \phi,
\]

which straightforwardly flows into

\[
\left( \frac{E_x}{E_{0x}} \right)^2 + \left( \frac{E_y}{E_{0y}} \right)^2 - 2 \left( \frac{E_x}{E_{0x}} \right) \left( \frac{E_y}{E_{0y}} \right) \cos \phi = \sin^2 \phi.
\]

Eq. 2.13 is the very equation of an ellipse which means the most general case of polarisation state is the elliptical one (see Fig. 2.2a). The other two polarisation states are special cases of the elliptical state.

Under the conditions of \( E_{0x} = E_{0y} \) and \( \phi = \pi/2 + \pi m \) (\( m \) is integer), Eq. 2.13 transforms into equation of a circle. Viewing from the direction towards which the wave is propagating, \( \phi = \pi/2 + 2\pi m \) corresponds to the electric field rotating clockwisely at a fixed \( z \) point, and the light is said to be right-handed circularly polarised (RCP) (see Fig. 2.2b). Considering the same viewing geometry, \( \phi = \pi/2 + 2\pi m \) corresponds to the electric field rotating counterclockwisely at a fixed \( z \) point, and the light is said to be left-handed circularly polarised (LCP) (see Fig. 2.2c).
Chapter 2. Theoretical Background

Figure 2.2. Schematic drawing of the polarisation states: a) elliptical state of polarisation, b) left-handed circular polarisation state, c) right-handed circular polarisation state, d) linear state of polarisation. The following notations are used: $E_{0x}$ and $E_{0y}$ are the electric field amplitudes along $x$- and $y$-axes, $\theta$ is angle between the major axis of the ellipse and $x$-axis, $\psi$ is angle of ellipticity and $\xi$ is angle between electric field vector and $x$-axis.

$-\pi/2 + 2\pi m$ corresponds to the electric field rotating counterclockwisely at a fixed $z$ point, and the light is said to be left-handed circularly polarised (LCP) (see Fig. 2.2b).\footnote{Note that definitions of right- and left-circular polarisations are just a matter of convention. We stick to the convention adopted by Azzam and Bashara \cite{5}.}

The case of $\phi = 0$ or $\pi m$ make Eq. \ref{eq:2.13} transform to give linear state of polarisation (see Fig. 2.2d).
2.2 The Jones Calculus

R. C. Jones [34] invented another deeply convenient method for representing the states of polarization.

Consider the same two orthogonal disturbances defined in Eqs 2.10 and 2.11 yet in scalar. A column vector, Jones vector, may be introduced to denote polarization state of a wave

\[
\mathbf{J} = \begin{bmatrix} E_x \\ E_y \end{bmatrix} = \begin{bmatrix} E_{0x} e^{i\phi_x} \\ E_{0y} e^{i\phi_y} \end{bmatrix},
\]

which represents a general case of elliptical polarization.

The two remaining basic polarization states may now be revisited using the concept of Jones vectors.

Linear polarization: keeping in mind \( \phi = \varphi_y - \varphi_x = 0 \) or \( \pi m \) and dividing Eq. 2.14 by the multiplication of \( E = (E_{0x}^2 + E_{0y}^2)^{1/2} \) and \( \exp(i\varphi_x) \), one arrives at

\[
\mathbf{J} = \begin{bmatrix} \cos \xi \\ \sin \xi \end{bmatrix},
\]

where \( \xi \) is an angle between the vector of electric field and \( x \)-axis.

The Jones vectors corresponding to RCP and LCP states can be obtained in the same way. The output takes on the following form:

\[
\mathbf{J}_R = \frac{1}{\sqrt{2}} \begin{bmatrix} 1 \\ i \end{bmatrix}, \quad \mathbf{J}_L = \frac{1}{\sqrt{2}} \begin{bmatrix} 1 \\ -i \end{bmatrix}.
\]

The whole elegance of the concept may be realized when one finds out it is possible to describe transformation of a polarization state after the light has passed through an element of an optical system. A 2 \( \times \) 2 matrix describing such a transformation may be assigned to that element. A number of elements in an optical system can be represented by a successive multiplication of the matrices corresponding to each of the elements. If the input Jones vector is given, the Jones 2 \( \times \) 2 matrix algebra provides the knowledge of polarization state at the exit of the optical system.

2.3 4-by-4 Matrix Formalism

Originally invented by Yeh [69] and further developed by Višňovský [64], the 4 \( \times \) 4 matrix formalism represents the amalgamation of Jones’ 2 \( \times \) 2 matrix method with the 2 \( \times \) 2 matrix method of Abelès [1].

Consider a multilayered structure in one dimension (1D) composed of \( I \) magnetic layers between two semi-infinite media. For an \( i \)th layer, Eq. 2.3 may be reformulated as:
Chapter 2. Theoretical Background

\[
\begin{bmatrix}
\frac{\partial^2 E^{(i)}}{\partial x^2} \\
\frac{\partial^2 E^{(i)}}{\partial y^2} \\
\frac{\partial^2 E^{(i)}}{\partial z^2}
\end{bmatrix} = \frac{1}{\varepsilon^2} \begin{bmatrix}
\varepsilon_{xx}^{(i)} & \varepsilon_{xy}^{(i)} & 0 \\
-\varepsilon_{xy}^{(i)} & \varepsilon_{yy}^{(i)} & 0 \\
0 & 0 & \varepsilon_{zz}^{(i)}
\end{bmatrix} \begin{bmatrix}
\frac{\partial^2 E^{(i)}}{\partial x^2} \\
\frac{\partial^2 E^{(i)}}{\partial y^2} \\
\frac{\partial^2 E^{(i)}}{\partial z^2}
\end{bmatrix}.
\] (2.17)

Substituting Eq. 2.5 into Eq. 2.17 and taking into account
\[\gamma^{(i)} = \pm (\omega/c)N^{(i)},\] (2.18)
where the sign \(\pm\) corresponds to forward- and backward-propagating waves, respectively, one may arrive at the following system of equations:

\[
\begin{bmatrix}
\varepsilon_{xx}^{(i)} - [N^{(i)}]^2 & \varepsilon_{xy}^{(i)} & 0 \\
-\varepsilon_{xy}^{(i)} & \varepsilon_{yy}^{(i)} - [N^{(i)}]^2 & 0 \\
0 & 0 & \varepsilon_{zz}^{(i)}
\end{bmatrix} \begin{bmatrix}
E_{0x}^{(i)} \\
E_{0y}^{(i)} \\
E_{0z}^{(i)}
\end{bmatrix} = 0.
\] (2.19)

The solution of the system results in
\[N^{(i)}_{\pm} = n^{(i)}_{\pm} - ik^{(i)}_{\pm} = (\varepsilon_{xx}^{(i)} \pm \varepsilon_{xy}^{(i)})^{\frac{1}{2}},\] (2.20)
where \(N\) denotes complex refractive index and the subscript \(\pm\), the correspondence to the \(\pm\) sign in front of \(\varepsilon_{xy}^{(i)}\). Eq. 2.20 shows there are two optical modes with \(N_+\) and \(N_-\) for forward propagation and two modes with the same \(N_+\) and \(N_-\) but propagating backwards. These four modes are associated with the two pairs of RCP and LCP modes.

The relations for total electric and magnetic fields in the \(i\)th layer and boundary conditions at the boundary between the \((i-1)\)th and \(i\)th layers for the same quantities bring one to the definition of the \(T^{(i-1,i)}\) matrix [64]. Such a matrix relates the electric field amplitudes at the boundary of the \((i-1)\)th and \(i\)th layers in the \((i-1)\)th layer and at the opposite boundary in the \(i\)th layer. \(T^{(i-1,i)}\) is defined as:

\[
T^{(i-1,i)} = \begin{bmatrix}
T_{11}^{(i-1,i)} & T_{12}^{(i-1,i)} & 0 & 0 \\
T_{12}^{(i-1,i)} & T_{22}^{(i-1,i)} & 0 & 0 \\
0 & 0 & T_{33}^{(i-1,i)} & T_{34}^{(i-1,i)} \\
0 & 0 & T_{43}^{(i-1,i)} & T_{44}^{(i-1,i)}
\end{bmatrix}.
\] (2.21)

where
2.3. 4-by-4 Matrix Formalism

\[ T_{11}^{(i-1,i)} = \frac{1}{2N_+(i-1)} (N_+^{(i-1)} + N_+^{(i)}) e^{i\beta_+^{(i)}}, \]  
\[ T_{12}^{(i-1,i)} = \frac{1}{2N_+(i-1)} (N_+^{(i-1)} - N_+^{(i)}) e^{-i\beta_+^{(i)}}, \]  
\[ T_{21}^{(i-1,i)} = \frac{1}{2N_+(i-1)} (N_+^{(i-1)} - N_+^{(i)}) e^{i\beta_+^{(i)}}, \]  
\[ T_{22}^{(i-1,i)} = \frac{1}{2N_+(i-1)} (N_+^{(i-1)} + N_+^{(i)}) e^{i\beta_+^{(i)}}, \]  
\[ T_{33}^{(i-1,i)} = \frac{1}{2N_-(i-1)} (N_-^{(i-1)} + N_-^{(i)}) e^{i\beta_-^{(i)}}, \]  
\[ T_{34}^{(i-1,i)} = \frac{1}{2N_-(i-1)} (N_-^{(i-1)} - N_-^{(i)}) e^{-i\beta_-^{(i)}}, \]  
\[ T_{43}^{(i-1,i)} = \frac{1}{2N_-(i-1)} (N_-^{(i-1)} - N_-^{(i)}) e^{i\beta_-^{(i)}}, \]  
\[ T_{44}^{(i-1,i)} = \frac{1}{2N_-(i-1)} (N_-^{(i-1)} + N_-^{(i)}) e^{-i\beta_-^{(i)}}. \]  

with \( \beta_+^{(i)} = (\omega/c) N_+^{(i)} d^{(i)} \) and \( d^{(i)} \), thickness of the \( i \)th layer.

The matrix \( M \) describes optical and MO response of the whole multilayer.

Thus, transmittance \( T \) and Faraday rotation \( \Theta_F \) of the multilayer may be calculated as

\[ T = \frac{s}{2} \left( |M_{11}|^2 + |M_{33}|^2 \right), \]  
\[ \Theta_F = -\frac{1}{2} \arg \left( \frac{M_{11}}{M_{33}} \right), \]

where \( s \) represents refractive index of a substrate.
2.4 Optical and Magneto-Optical Characteristics for a Single Layer

Transmittance

Off-diagonal element of the permittivity tensor $\varepsilon_{xy}$ is much less than the diagonal one $\varepsilon_{xx}$. It, therefore, may be neglected when transmittance is considered and for a single film Eq. 2.31 may be simplified to give the Swanepoel formula [60]:

$$T = \frac{1 + s^2}{2s} \frac{Ax}{B - Cx + Dx^2}, \quad (2.33)$$

where

$$A = 16s(n^2 + k^2),$$
$$B = [(n + 1)^2 + k^2]([(n + 1)(n + s^2) + k^2],$$
$$C = [(n^2 - 1 + k^2)(n^2 - s^2 + k^2) - 2k^2(s^2 + 1)]2 \cos \phi$$
$$-k[2(n^2 - s^2 + k^2) + (s^2 + 1)(n^2 - 1 + k^2)]2 \sin \phi,$$
$$D = [(n - 1)^2 + k^2][(n - 1)(n - s^2) + k^2],$$
$$\phi = 2\gamma nd,$$
$$x = \exp(-2\gamma kd).$$

Here $n$, $k$ and $d$ are refractive index, extinction coefficient and thickness of a film, respectively, $s$, refractive index of a substrate.

Faraday Rotation

The case of a single film makes Eq. 2.32 become simplified and one may arrive at the following equation for FR [63]:

$$\Theta_F = \Re \left\{ \frac{N_+ - N_-}{4N} \cdot \frac{2\gamma Nd(1 + r_{fa}r_{fs}e^{-i2\gamma Nd}) + i(e^{-i2\gamma Nd} - 1)(r_{fa} + r_{fs})}{1 - r_{fa}r_{fs}e^{-i2\gamma Nd}} \right\}, \quad (2.34)$$

where $r_{fa} = \frac{N - 1}{N + 1}$ is reflection coefficient at the air-film interface, $r_{fs} = \frac{N - s}{N + s}$ is reflection coefficient at the film-substrate interface, and $N = \sqrt{\varepsilon_{xx}}$ represents complex refractive index of a film from Eq. 2.20 with $\varepsilon_{xy} = 0$. 

3.1 Iron Garnets

The crystalline structure of iron garnets corresponds to a slightly distorted body-centered cube with a fairly large lattice constant (1.2 – 1.3 nm) compared with typical solids. This structure consists of the three types of crystallographic sites: dodecahedral, octahedral, and tetrahedral. Here the metal ions have eight, six and four oxygen ions in the neighbourhood, respectively [41].

The structure of iron garnets gives rise to a large magnetic anisotropy which leads to occurrence of magneto-optical activity. Such an activity in the form of the Faraday rotation has found an application to optical isolators. Thin films of pure iron garnets, however, provide just moderate magnitude of Faraday rotation. The trends to miniaturisation of components in optical systems bring about a demand for reduction of dimensions of active as well as passive elements significantly. Owing to this reason, there were a lot of attempts to enhance Faraday rotation in thin films of iron garnets. This was done by substitution into the three sites of the iron garnet structure by other metals.

Almost all pure iron garnets were investigated in terms of optical and magneto-optical properties. Several exceptions include Pr$_3$Fe$_5$O$_{12}$, Nd$_3$Fe$_5$O$_{12}$, Ce$_3$Fe$_5$O$_{12}$ and Pm$_3$Fe$_5$O$_{12}$. A rigorous literature search revealed few papers devoted to the study of magnetic properties of Pr$_3$Fe$_5$O$_{12}$ and Nd$_3$Fe$_5$O$_{12}$ (see e.g. [59]). No information was found concerning Ce$_3$Fe$_5$O$_{12}$ and Pm$_3$Fe$_5$O$_{12}$. The latter can hardly be examined in view of its radioactivity. A fact to be mentioned is that the properties of pure iron garnets exhibit close resemblance because of similarity in the environment of the iron ions [30].

Three chief substitutions into the octahedral and tetrahedral sublattices were carried out for some IGs: YIG:Ga [59], DyIG:Sc [54], HoIG:Sc [51], ErIG:Sc [52], ErIG:Al [50] and YbIG:Sc [53]. Al and Ga were found to have a preference for substitution into the tetrahedral sublattice, whereas Sc enters the octahedral one. The so-called dilution effect taking place on substitution with the three metals leads
a drop in the magneto-optical activity [30].

Several iron garnets were used as starting materials for substitutions into the dodecahedral sublattice: \( \text{Y}_3\text{Fe}_5\text{O}_{12} \) (YIG) in which the majority of substituents were incorporated, \( \text{Gd}_3\text{Fe}_5\text{O}_{12} \) (GdIG), TmIG, YbIG and several others. Buhrer [8] was the first who discovered the substitution of rare earth (RE) in RE iron garnets for Bi results in much larger Faraday rotation values in comparison with the pure garnet systems. An enhancement of Faraday rotation similar to Bi but lower in magnitude was also found for Ce [21], Pb [30] and Pr [22]. A body of attempts were directed to obtaining completely substituted YIG by Bi. Okuda et al. first reported on successful deposition of bismuth iron garnet \( \text{Bi}_3\text{Fe}_5\text{O}_{12} \) (BIG) by reactive ion beam sputtering [49]. Rapid development of pulsed laser deposition (PLD) also enabled deposition of BIG [2].

BIG provides the record values of specific FR, e.g. \( \theta_F = -8.4^{\circ}/\mu\text{m} \) at \( \lambda = 633\text{ nm} \). The question about the reasons for such a dramatic increase in Faraday rotation remains to be answered. A reasonable speculation may be a large ionic radius of Bi which is the rationale for large distortions of the lattice as incorporation of Bi takes place. The lattice distortions result in enhancement of superexchange interactions as well as spin-orbit coupling between the ions of the unit cell. This, in turn, have the prime impact on the behaviour of FR.

There is one intrinsic problem of BIG: it is fairly opaque in visible light due to an absorption edge at around 550 nm. The strong correlation between absorption and Faraday rotation spectra makes it difficult for BIG to find industrial implementations. The largest Faraday rotation, therefore, has to be traded off against absorption which may be realised in 2 ways. The first way means turning back to substituted systems. Some advanced compositions, viz. Ce-containing iron garnets [31], \( \text{YYbBiIG} \) [32] and \( \text{TbYbBiIG} \) [73], have already shown a substantial potential for operation in the near and short-wavelength infrared (IR) spectral ranges. The second approach consists in construction of PCs [36, 38, 39]. The spectral range of visible light still represents a formidable challenge.

### 3.2 Titanium Dioxide

Titanium oxide \( \text{TiO}_2 \) or titania has also been drawing a deal of attention from academic as well as industrial viewpoints. Unique properties of titania hold a prime impact on established applicability of this material.

Metal/oxide structures represent rather attractive alternatives to bulk metal catalysts in many industrial processes. A metal put onto oxide support not only decreases the cost of the catalyst but also improves catalytic activity of the metal. The case of \( \text{TiO}_2 \) is of particular significance since several metals, such as Ni, enhance the reaction kinetics drastically [5]. Titania, however, does not act as a suitable oxide support due to 'strong metal-support interaction'. This effect leads to encapsulation of metal clusters by titanium suboxides and decrease of catalytic activity [55]. 2D and 3D islands of gold on \( \text{TiO}_2 \) (110) are exempt from the encap-
3.2. Titanium Dioxide

sulation and represent a promising catalyst for room temperature oxidation of CO

UV illumination of titanium dioxide results in creation of electron-hole pairs. The charge carriers produced partake in reactions with the absorbed molecules on the surface of TiO$_2$. The resulting radical species possess profound reactivity to decomposition of organic molecules. This fact was at once recognised to be of great importance in purification of water [12], disinfection [15], self-cleaning and antifogging coatings [70].

Thin films of TiO$_2$ change their conductance upon exposure with several gases. This effect has found its application to gas sensing. The results of successful fabrication and tests of sensors for monitoring of humidity [10] as well as detection of ammonia [37] and oxygen [58] were published.

The complementary metal-oxide-semiconductor as well as related technologies pose new challenges of decreasing device feature sizes. Size shrinkage of the gate dielectric is becoming vital and silicon oxide will soon no longer meet the requirements in this respect. The need for a novel material capable of replacing SiO$_2$ has emerged. TiO$_2$ is regarded as a potentially suitable alternative for the replacement in view of its high dielectric constant (80 - 110 depending on the deposition method). In spite of its high permittivity, titania as a layer in transistors and capacitors is responsible for two deleterious processes: formation of reduced titanium oxide landing in carrier trapping and high leakage paths, as well as uncontrolled formation of interfacial SiO$_2$ layer [35, 66].

Among the properties of titanium dioxide, a special emphasis is to be put on its optical properties. Existing in three major crystalline phases — rutile (tetragonal, $a = b = 4.584$ Å, $c = 2.953$ Å), anatase (tetragonal, $a = b = 3.782$ Å, $c = 9.502$ Å) and brookite (rhombohedral, $a = 5.436$ Å, $b = 9.166$ Å, $c = 5.135$ Å) [16] — TiO$_2$ owns one of the highest refractive indices (2.72 at 550 nm for a rutile single crystal) [24]. In addition, TiO$_2$ is transparent in the visible light which makes it a useful material for various applications in optics [57].

Just anatase and rutile phases are available in thin films [42] having lower refractive indices in comparison with their bulk analogues. A great body of deposition methods were utilised to obtain thin films of titania with the refractive indices and absorption (extinction) coefficients close to TiO$_2$ in bulk. Table 3.1 gives a glimpse at the film characteristics provided by different deposition techniques. The root-mean-square (RMS) roughness is also given since interfacial roughness has a crucial influence on transparency of films.

The table presents the best characteristics of titania films obtained by a deposition technique so far. Apparently, physical methods of deposition provide higher refractive index values than chemical ones. Among physical deposition methods, the best results were found for the case of filtered cathodic vacuum arc. PLD comes close a second and provides completely acceptable characteristics of TiO$_2$ for being high refractive index material in PCs.

Because of the interest in manufacture of photonic crystals exhibiting photoluminescence, the search was performed for a material exhibiting luminescence and
lower refractive index in comparison with TiO$_2$. A material satisfying the requirements is erbium oxide.
Table 3.1. Comparison of optical characteristics of TiO\textsubscript{2} obtained by different deposition techniques.

<table>
<thead>
<tr>
<th>Deposition technique\textsuperscript{a}</th>
<th>$T_s$ [°C]</th>
<th>$T_{an}$/time [°C/h]\textsuperscript{b}</th>
<th>Crystalline phase</th>
<th>$n$</th>
<th>$k$ at $\lambda = 550$ nm</th>
<th>RMS roughness</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sol-gel spin coating\textsuperscript{[65]}</td>
<td>Room</td>
<td>350/1</td>
<td>Anatase</td>
<td>2.3</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Sol-gel dip coating\textsuperscript{[11]}</td>
<td>Room</td>
<td>600/3</td>
<td>Anatase</td>
<td>2.44</td>
<td>-</td>
<td>0.9</td>
</tr>
<tr>
<td>DC magnetron sputtering\textsuperscript{[43]}</td>
<td>Room</td>
<td>-</td>
<td>Anatase/rutile</td>
<td>2.62</td>
<td>$2.5 \cdot 10^{-3}$</td>
<td>19.45</td>
</tr>
<tr>
<td>Reactive RF sputtering\textsuperscript{[44]}</td>
<td>250</td>
<td>-</td>
<td>Anatase/rutile</td>
<td>2.57</td>
<td>$4.0 \cdot 10^{-3}$</td>
<td>5.5</td>
</tr>
<tr>
<td>DC magnetron sputtering with ion beam assistance\textsuperscript{[40]}</td>
<td>75</td>
<td>500/1</td>
<td>Anatase</td>
<td>2.52</td>
<td>$4.2 \cdot 10^{-3}$</td>
<td>1.5-2.6</td>
</tr>
<tr>
<td>Pulsed bias arc ion plating\textsuperscript{[72]}</td>
<td>Room</td>
<td>-</td>
<td>Amorphous</td>
<td>2.51</td>
<td>-</td>
<td>0.11</td>
</tr>
<tr>
<td>PLD\textsuperscript{[18]}</td>
<td>150</td>
<td>500/30 [°C/s]</td>
<td>Anatase</td>
<td>2.53</td>
<td>$2.0 \cdot 10^{-3}$</td>
<td>0.6</td>
</tr>
<tr>
<td>Filtered cathodic vacuum arc\textsuperscript{[74]}</td>
<td>Room</td>
<td>-</td>
<td>Amorphous</td>
<td>2.56</td>
<td>$1.0 \cdot 10^{-4}$</td>
<td>0.5</td>
</tr>
</tbody>
</table>

\textsuperscript{a}The techniques are listed in the order of enhancement of optical performance

\textsuperscript{b}Post-annealing, if performed
3.3 Erbium Oxide

Erbium oxide \( \text{Er}_2\text{O}_3 \) is the active component of optical amplifiers used for data transfer for telecommunication purposes. The interest in it originally came out in 1987 when the first optical fibre amplifier was made [47]. This circumstance stimulated active research on optical properties of Er-doped media including semiconductors, dielectrics and ceramics. The studies at issue comprise first of all investigations of luminescence spectra, lifetimes, concentration quenching and upconversion.

In accordance with the National Institute of Standards and Technology, a free ion of \( \text{Er}^{3+} \) exhibits 9 spectral lines. The corresponding energy levels are schematically depicted in Fig. 3.1

Owing to the Stark effect, the energy level diagram of \( \text{Er}^{3+} \) ions in compounds becomes more involved. The extent of the complexity depends on crystalline environment of the ions. A single crystal of \( \text{Er}_2\text{O}_3 \), for instance, possesses around 600 spectral lines [29].

Just few levels are available whose lifetimes are sufficient to cause radiative transitions. In Er-doped glasses, \( ^4I_{13/2} \rightarrow ^4I_{15/2} \) is a radiative transition at room temperature lasing at around 1.54 \( \mu \text{m} \). The lifetime on the metastable \( ^4I_{13/2} \) level is in the order of 10 ms depending on the host glass composition. Such a large lifetime allows substantial population inversion and leads to high gain and low noise using low power pumping [17]. Presence of Stark sublevels belonging to the \( ^4I_{13/2} \) and \( ^4I_{15/2} \) levels brings about appearance of additional spectral lines in the luminescence spectra in the region \( 1.48 - 1.63 \mu \text{m} \).

There are two more transitions that have also been investigated extensively. The first transition \( ^4I_{11/2} \rightarrow ^4I_{13/2} \) provides emission at around 2.7 \( \mu \text{m} \). The transition is called self-terminating in that the \( ^4I_{13/2} \) level has longer lifetime in comparison with the \( ^4I_{11/2} \) one. The second one \( ^4I_{11/2} \rightarrow ^4I_{15/2} \) lases at about 0.98 \( \mu \text{m} \). Depending on the host glass composition, the lifetimes on the \( ^4I_{11/2} \) level vary but do not exceed 10 ms [17].

Concentration quenching manifests itself in reduction of quantum efficiency of ions when the ion concentration is increased. The result of increasing ion concentration is that the ions tend to form clusters which enhances interionic interactions. Experimental studies revealed significant drop in quantum efficiency for Er-doped silica fibres at Er concentrations higher than \( 10^{18} \text{ cm}^{-3} \) [15]. Co-doping with Ge and Al sets a higher limit of quenching onset [35] by reducing the tendency for Er to clustering. Another deleterious process is the energy transfer to \( \text{OH}^- \)-groups that act as excellent traps at low Er concentrations [20]. Annealing was proven to be a suitable remedy to removal of \( \text{OH}^- \)-groups [19].

There are two mechanisms responsible for upconversion: sequential absorption of pump photons by excited-state absorption and energy transfer between two excited ions. Both of the processes are found in Er-doped glasses. The first one arises with pumping at 0.8 \( \mu \text{m} \) where successive absorption of two photons takes place. At high-power pumping, the two-photon absorption results in \( ^2H_{9/2} \rightarrow ^4I_{15/2} \) transition and corresponding emission of a photon with \( \lambda \approx 0.4 \mu \text{m} \). The high probability
of a non-radiative transition from the $^2H_{9/2}$ level accounts for the luminescence at about 0.55 $\mu$m that issues from the $^4S_{3/2}$ level. The population of the $^4F_{9/2}$ level by non-radiative relaxation from $^4S_{3/2}$ is also highly probable giving emission at around 0.66 $\mu$m [51]. In general, upconversion is regarded as a parasitic process, for it reduces luminescence efficiency. The phenomenon is being studied vigorously nowadays in view of its applicability to fabrication of upconversion lasers (see e.g. [3, 56]).
Chapter 4

Characterisation Techniques

4.1 X-Ray Diffraction

X-ray diffraction (XRD) may be regarded as constructive interference of reflected X-ray beams. From resultant diffraction pattern, it is possible to extract a detailed information on the structure and crystalline phase of a sample.

Assuming elastic scattering, the reflected wave will acquire a wave vector $\gamma'$ (see Fig. 4.1). The necessary conditions for occurrence of a diffracted beam are represented by the Laue equations

$$a \cdot (\gamma' - \gamma) = 2\pi q,$$
$$b \cdot (\gamma' - \gamma) = 2\pi p,$$
$$c \cdot (\gamma' - \gamma) = 2\pi v,$$

(4.1)

where $q$, $p$ and $v$ are integral numbers, and $a$, $b$ and $c$ are the lattice unit vectors. The Laue equations have a simple geometrical interpretation. If one defines the reciprocal lattice vector

$$G = hH + kK + lL,$$

(4.2)

where $h$, $k$ and $l$ are the Miller indices and $H$, $K$ and $L$ are the fundamental vectors of the reciprocal lattice, the diffraction condition applies if

$$\gamma' - \gamma = G.$$

(4.3)

In other words, $\gamma'$, $\gamma$ and $G$ have to form a triangle in the reciprocal space (so-called Ewald construction) as it is shown in Fig. 4.1.

The well-known Bragg law may be derived from the Laue equations. The law describes the occurrence of interference maxima from simple geometrical considerations: $m\lambda = 2d\sin \theta$, with $m$ being integer, $\lambda$, wavelength of X-rays, $d$, the distance

\[\text{For the full derivation of the concept, the reader is warmly welcome to consult Ref. 14.} \]
between the nearby atomic planes and $\theta$, angle of incidence. The law is particularly useful for extraction of the interplane distance $d$.

The Siemens D5000 powder diffractometer was employed to collect XRD patterns of samples in the $\theta-2\theta$ scan mode. The following settings of the diffractometer were used: Cu $K_{\alpha 1}$ radiation with $\lambda = 1.54056$ Å, a voltage of 40 kV, a current of 30 mA, and a 0.01° step size with a time of 7 sec at each step. The XRD analysis was performed with the aim of obtaining information on the presence of crystalline phases in the samples.

4.2 Profilometry

Profilometry represents a widely-used technique for imaging surface profiles. It is also one of the easiest ways of measuring film thickness.

A diamond stylus brought in contact with a sample surface detects the surface features by changing its vertical position. An analog signal created thereby becomes then converted into a digital one. The digital signal serves as a means of reconstruction of the surface profile.

For measurements of film thickness, several parts of a substrate, typically its corners, should be covered by a soft mask. After deposition, the mask is removed releasing steps of the substrate corners and the film. The stylus moved over the step gives film thickness. In spite of the apparent ease of use, profilometry possesses a significant drawback: film thickness can be measured only in one particular direction. In order to produce a reliable value of film thickness, the measurements have to be conducted in several different directions on each corner. The ultimate value of the thickness is found as an average over all of the values.
4.3 Atomic Force Microscopy

The up-to-date instruments are capable of producing surface maps which eliminates the need for performing measurements in different directions.

Thicknesses of the films were measured by the Alpha Step 200 profilometer with a stylus diameter of 12.5 µm and a shank angle of 60°. The scan length was set to 400 µm and sampling rate to 50 µm/sec.

4.3 Atomic Force Microscopy

A much more sophisticated method of investigation of surface topography is atomic force microscopy (AFM). Apart from imaging surface profiles, AFM is highly utilisable for measurements of forces including van der Waals, electrostatic, magnetic, adhesion forces and such. In contrast to other well-established techniques, it does not require any special way of sample preparation and provides a possibility of conducting measurements in fluid media.

The operational principle of AFM may be illustrated as follows. A laser beam is reflected off the back of a cantilever. Upon the reflection, detection of the laser beam takes place by a split photodetector of subangstrom sensitivity. To preclude a crash of the cantilever against sample surface, a piezoscanner and a feedback loop adjust $z$-position of the cantilever.

There are two operational modes of AFM differing in mechanism of signal detection. In contact mode, the feedback loop adjusts the $z$-position of the cantilever by constant monitoring of the tip-sample force. The variation in the $z$-position represents a signal recorded. In dynamic mode which includes tapping and non-contact modes, the cantilever is forced to oscillate close to its resonance frequency. When the cantilever approaches the surface, the oscillations become damped. The oscillation amplitude decreases which serves as a signal detected.

In all of the experiments, the Nanoscope III instrument was used to collect the AFM images. The tip was made of highly doped silicon with a radius of curvature less than 10 nm. The scans were made in tapping mode over an area of $2 \times 2 \mu m^2$.

4.4 Optical Spectrometry

Fourier transform infrared spectrometry (FTIR) is a well-established means of recording optical spectra.

The operational principle of a FTIR spectrometer is based on the following. Upon emission from a source, a light beam is reflected off the mirror and having passed through a beam splitter becomes divided into two parts. One part comes into interferometer that represents a compartment of the spectrometer with a moving mirror. The mirror is used to create a path difference $x$. After reflection inside the interferometer, the beam part recombines with the other part and both of them pass through the sample and become detected. The software of the spectrometer constructs an interferogram from which the intensity is calculated.
Chapter 4. Characterisation Techniques

The mathematics of the software lying behind is based on interference algebra. Consider two plane waves. Interference of the waves allows writing the resultant intensity as

\[ I = E_1^2 + E_2^2 + 2E_1E_2 \cos \phi. \] (4.4)

For normal incidence \( \phi = \gamma x \) and Eq. 4.4 transforms to

\[ I(x) = 2[1 + \cos(\gamma x)], \] (4.5)

which becomes

\[ I(x) = \int_0^\infty [1 + \cos(\gamma x)] \Im(\gamma) d\gamma \] (4.6)

for non-monochromatic light.

Eq. 4.6 may be simplified to the following form:

\[ I(x) = \frac{1}{2} I(0) + \frac{1}{2} \int_{-\infty}^{\infty} \Im(\gamma)e^{i\gamma x} d\gamma, \] (4.7)

consequently,

\[ F(x) = \frac{2I(x) - I(0)}{\sqrt{2\pi}} = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} \Im(\gamma)e^{i\gamma x} d\gamma \] (4.8)

represents a Fourier transform. This means that the detected intensity \( I(x) \) as a function of position of moving mirror can be converted into \( \Im(\gamma) \) that is an intensity spectrum as a function of frequency by the Fourier transform.

In order to collect transmittance spectra of the films in a range \( 0.4 - 2.6 \mu m \), the Nicolet 5700 FTIR spectrometer was used. The normalisation procedure was done to bare substrates. There is one detail needing to be noted. All of the transmittance spectra recorded lack data points in the vicinity of 632 nm. The reason for such a circumstance is the laser operating at \( \lambda = 632 \) nm that is used for internal calibration of the instrument.
Part II

Original Results
Chapter 5

Optical Properties of Titanium Dioxide

All of the TiO$_2$ films were deposited by pulsed laser deposition system ‘Lambda Physik LPX 305iCC’. Parameters of the deposition are presented in Table 5.1.

There are three foremost quantities to pursue whilst considering optical performance of thin films: maximal refractive index, lowest absorption and lowest surface roughness. Amorphous films are known to have extremely low surface roughnesses. The choice of the substrate temperature $T_s = 150 \degree C$ resulted in an RMS surface roughness $\sigma$ of TiO$_2$ films in the order of 0.4 nm which is very promising. However, transmittance spectra of the as-deposited films possess a drop of transmittance envelope centered at 0.9 $\mu m$ (see Fig. 5.1). Thus, the requirement for the lowest absorption is not satisfied. In order to dispose of the drop, reach maximal refractive index and preserve low surface roughness, we employ three routines of tailoring optical properties of TiO$_2$ films. These are deposition at elevated $T_s$, regular annealing in thermodynamically equilibrium conditions, and rapid thermal annealing (RTA).

$^1$Occurrence of the drop might be attributed to the presence of oxygen vacancies and Ti$^{3+}$ crystal field transitions [7, 23].

**Table 5.1.** Parameters of pulsed laser deposition process of titania films.

<table>
<thead>
<tr>
<th>Settings of laser</th>
<th>Description</th>
<th>Process parameters</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>Wavelength</td>
<td>248 nm</td>
<td>Target</td>
<td>TiO$_2$ (99.9 %)</td>
</tr>
<tr>
<td>Pulse width</td>
<td>25 ns</td>
<td>Substrate</td>
<td>SiO$_2$ (Corning glass 7059)</td>
</tr>
<tr>
<td>Energy density</td>
<td>300 mJ/cm</td>
<td>Target-to-substrate distance</td>
<td>8.5 cm</td>
</tr>
<tr>
<td>Repetition rate</td>
<td>30 Hz</td>
<td>Initial substrate temperature</td>
<td>150 $^\circ$C</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Base pressure</td>
<td>$&lt; 4.0 \cdot 10^{-5}$ mbar</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Oxygen pressure</td>
<td>$1.4 \cdot 10^{-2}$ mbar</td>
</tr>
</tbody>
</table>
Chapter 5. Optical Properties of Titanium Dioxide

Figure 5.1. Transmittance spectra of the titania films: T150, a spectrum of the film deposited at 150 °C; a) T300 and T500: the spectra of the films deposited at 300 and 500 °C, respectively; b) A300 and A800: the spectra of the films post-annealed at 300 and 800 °C, respectively; c) RTA500 and RTA800: the spectra of the films subjected to rapid thermal annealing at 500 and 800 °C. The spectra are depicted using the energy scale $1/\lambda$. 
5.1 Deposition at Elevated Substrate Temperatures

Depositions at elevated $T_s$ were performed at 300, 400 and 500 $^\circ$C. The corresponding samples are denoted by adding the letter T in front of the temperatures, e.g. T300 stands for the sample deposited at $T_s = 300$ $^\circ$C.

Fig 5.1a shows transmittance spectra of the films deposited at elevated $T_s$. To avoid confusion, the transmittance spectra of only T300 and T500 samples are depicted. Clearly, the drop vanishes at $T_s = 300$ $^\circ$C showing appropriate values of refractive index $n$, absorption coefficient $\alpha$ and RMS surface roughness $\sigma$ (see Table 5.2). With the increase in $T_s$, a rise in $\sigma$ as well as $n$ takes place. A reasonable conjecture is a growing extent of crystallinity which was proved by XRD. The T300 and T400 samples were found to have the crystalline structure of anatase and T500 sample was composed of a mixture of anatase and rutile (Fig. 5.2, pattern As—deposited at 500 $^\circ$C). Rather high $\sigma$ of T500 sample is inappropriate regardless of such high $n$. The two other samples provide quite suitable values of $n$, $\alpha$ and $\sigma$.

5.2 Regular Annealing

The second routine was regular annealing. Originally deposited at 150 $^\circ$C, the films were post-annealed in air under thermodynamically equilibrium conditions. Post-annealing temperatures were chosen to be 300, 400, 500, 600, 700 and 800 $^\circ$C. The sample designation is A300, A400, etc. corresponding to the films annealed at 300, 400, etc. $^\circ$C. The rate of increase in post-annealing temperature was such that the set points (300, 400, ... $^\circ$C) were reached in 60 min for all of the samples except for the A700 and A800 ones. Their set points were reached in 70 and 80 min, correspondingly.

Annealing at 300 $^\circ$C eliminates the drop in transmittance of as-deposited films (Fig. 5.1b, spectrum A300) giving lower absorption in comparison with the T300-

In the following two routines, just the extremities, i.e. the samples subjected to lowest and highest temperatures of processing, shall be shown because of the same reason.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Deposition at different $T_s$</th>
<th>Regular annealing</th>
<th>RTA</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>T300</td>
<td>T400</td>
<td>T500</td>
</tr>
<tr>
<td>Structure</td>
<td>An</td>
<td>An</td>
<td>An/Ru</td>
</tr>
<tr>
<td>$n_{0.55}$</td>
<td>2.41</td>
<td>2.45</td>
<td>2.53</td>
</tr>
<tr>
<td>$\alpha_{0.55}$ [cm$^{-1}$]</td>
<td>4010</td>
<td>3890</td>
<td>4079</td>
</tr>
<tr>
<td>$\sigma$ [nm]</td>
<td>1.5</td>
<td>1.6</td>
<td>8.7</td>
</tr>
</tbody>
</table>
Figure 5.2. XRD patterns of TiO$_2$ films: Anatase JCPDS (●) and Rutile JCPDS (■), the 73-1764 and 78-1510 patterns of anatase and rutile, respectively, taken from the Joint Committee on Powder Diffraction Standards database; As-deposited at 500 °C, the sample deposited at 500 °C; Post-annealed at 800 °C, the sample deposited at 150 °C and post-annealed at 800 °C; RTA at 800 °C, the sample deposited at 150 °C and rapid-thermally annealed at 800 °C.

T500 samples (Table 5.2). A further increase in annealing temperature brings about larger absorption that reaches a value of 15024 cm$^{-1}$ for the sample A800. Hardly any change in $n$ suggests the films become optically dense and well-crystallised already during annealing at 300 °C. Analysis of XRD patterns of the samples evidences the presence of anatase phase after all of the annealing stages (Fig. 5.2, pattern Annealed at 800 °C). $\sigma$ does not variate significantly with the increase in annealing.
5.3. Rapid Thermal Annealing

RTA procedure was carried out in air at 500, 600, 700 and 800°C. The respective sample denotement is RTA500, RTA600, etc. The annealing times were set to 30 sec for RTA500 and 15 sec for the remaining ones. The annealing time of 30 sec for the case of the RTA500 sample was chosen since a shorter time did not produce any changes in film properties.

RTA in the whole range of temperatures made the drop disappear (Fig. 5.1c). The same trends as in regular annealing apply for the case of RTA. With the increase in annealing temperature, \( \alpha \) exhibits increase from 404 to 2500 cm\(^{-1}\) and \( \sigma \) from 0.6 to 1.2 nm for the RTA500 – RTA800 samples. \( n \) is found to be brought up slightly as well. The sole crystalline phase occurring after the RTA treatments is anatase (see Fig. 5.2, pattern RTA at 800°C).

Summary

Comparing optical performance of the TiO\(_2\) films in terms of the three routines summarised in Table 5.2, we conclude the best performance shows the film subjected to RTA at 500°C. Such a film possesses a refractive index \( n = 2.53 \) and an absorption coefficient \( \alpha = 404 \text{ cm}^{-1} \) at \( \lambda = 0.55 \mu\text{m} \), as well as an RMS surface roughness \( \sigma = 0.6 \text{ nm} \).
Chapter 6

Simulations of Transmittance and Faraday Rotation

6.1 Single Film Materials

For simulations of transmittance $T$ and the Faraday rotation $\Theta_F$ for single film materials, one may employ Eqs 2.33 and 2.34. In these equations, there are three quantities to be concerned with: refractive index $n$ and extinction coefficient $k$ of a film, as well as refractive index of a substrate $s$.

Single films as well as PCs were grown on two substrates: Corning glass 7059 (CG) and Ca-, Mg-, Zr-doped gadolinium gallium garnet Ca,Mg,Zr:Gd$_3$Ga$_5$O$_{12}$ (CMZGGG). The specification sheet of CG provides three values of refractive index: 1.5440 at 435.8 nm, 1.5354 at 546.1 nm and 1.5311 at 643.8 nm [13]. These values were fitted by the least square method to produce a wavelength dispersion of $s$. The dispersion obtained has a form of a single-term Sellmeier equation:

$$s_{CG}^2 = 1 + \frac{1.312}{1 - \left(\frac{0.099 \mu m}{\lambda}\right)^2}. \quad (6.1)$$

The refractive index of CMZGGG exhibits the values akin to Gd$_3$Ga$_5$O$_{12}$ (GGG). The dispersion relation for GGG is reported in [67]. It has 3 terms to account for the refractive index in a wavelength range from near ultraviolet to middle IR. The wavelength range of our interest is $0.38 - 2.63 \mu m$ which allows using a single-term Sellmeier equation

$$s_{CMZGGG}^2 = 1 + \frac{2.75}{1 - \left(\frac{0.128 \mu m}{\lambda}\right)^2}. \quad (6.2)$$

as dispersion of the refractive index for Ga,Mg,Zr,Gd$_3$Ga$_5$O$_{12}$.

In order to obtain $n$'s of TiO$_2$, Er$_2$O$_3$ and Bi$_3$Fe$_5$O$_{12}$ let us examine a transmittance spectrum of a film (Figs 6.1, 6.2, 6.3). The spectrum is made of Fabry-Pérot oscillations. The extrema positions of the oscillations are defined by
Chapter 6. Simulations of Transmittance and Faraday Rotation

Figure 6.1. The spectra of: a) transmittance of TiO$_2$ film, b) transmittance of Er$_2$O$_3$ film, c) transmittance of BIG film, d) Faraday rotation of BIG film. The symbol ○ denotes the experimental data and the line —, the results of the simulations.

A resonance condition $c \lambda_e = 2n(\lambda_e)d$. Here $c$ is an integer for maxima and half-integer for minima, $\lambda_e$ is wavelength at which the oscillation extremum appears, $n(\lambda_e)$ is refractive index of the film at $\lambda_e$ and $d$ is film thickness. After $\lambda_e$’s are extracted from the spectra, the values of $n(\lambda_e)$ can be calculated. The values obtained are then fitted by a single-term Sellmeier equation to produce the wavelength dispersions of refractive indices pertaining to TiO$_2$, Er$_2$O$_3$ and BIG:

\begin{align*}
n_{TiO_2}^2 &= 1 + \frac{4.40}{1 - \left(\frac{0.240 \text{ } \mu\text{m}}{\lambda}\right)^2}, \\
n_{Er_2O_3}^2 &= 1 + \frac{2.65}{1 - \left(\frac{0.150 \text{ } \mu\text{m}}{\lambda}\right)^2}, \\
n_{BIG}^2 &= 1 + \frac{4.92}{1 - \left(\frac{0.310 \text{ } \mu\text{m}}{\lambda}\right)^2}.\end{align*}  
(6.3)
(6.4)
(6.5)
Comparison of Eqs 6.3–6.5 with Eq. 2.8 enables extraction of the resonance angular frequencies $\omega_0$ and plasma frequencies multiplied by oscillator strengths $\omega^2_p f$ for the materials. These values are taken as the corresponding quantity values of the first short-wavelength dipole transition. To take account of resonance features in transmittance spectra of TiO$_2$, Er$_2$O$_3$ and BIG, several other dipole transitions were introduced (see Table 6.1).

The material parameters of TiO$_2$, Er$_2$O$_3$ and BIG give wavelength dispersions of their $n$, $k$ and $N$ (as well as $N_\pm$ for BIG). It is also possible to obtain the interpolation formulae for the complex refractive indices of the single film materials:

For TiO$_2$

\[
N^2_{\text{TiO}_2} = 1 + \frac{4.44}{1 - (\frac{0.238 \mu m}{\lambda})^2 + i \cdot 5.76 \cdot 10^{-3} \cdot (\frac{0.238 \mu m}{\lambda})^3 + 0.012} \left[1 - (\frac{1.11 \mu m}{\lambda})^2 + i \cdot 0.92 \cdot (\frac{1.11 \mu m}{\lambda})^3\right],
\] (6.6)

for Er$_2$O$_3$

\[\text{Table 6.1. Material parameters of TiO}_2$, Er$_2$O$_3$ and Bi$_3$Fe$_5$O$_{12}$.

<table>
<thead>
<tr>
<th>Transition no.</th>
<th>$\lambda_0$ [\mu m]</th>
<th>$\hbar \omega_0$ [eV]</th>
<th>$\hbar^2 \omega^2_p f$ [eV]</th>
<th>$\hbar \Gamma$ [eV]</th>
<th>$\hbar \Delta$ [eV]</th>
</tr>
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<tbody>
<tr>
<td><strong>TiO$_2$</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1</td>
<td>0.238</td>
<td>5.20</td>
<td>120</td>
<td>1.50 \cdot 10^{-2}</td>
<td>0</td>
</tr>
<tr>
<td>2</td>
<td>1.25</td>
<td>0.990</td>
<td>1.50 \cdot 10^{-2}</td>
<td>0.510</td>
<td>0</td>
</tr>
<tr>
<td><strong>Er$_2$O$_3$</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1</td>
<td>0.149</td>
<td>8.30</td>
<td>183</td>
<td>1.50 \cdot 10^{-2}</td>
<td>0</td>
</tr>
<tr>
<td>2</td>
<td>0.523</td>
<td>2.37</td>
<td>6.00 \cdot 10^{-4}</td>
<td>6.00 \cdot 10^{-3}</td>
<td>0</td>
</tr>
<tr>
<td>3</td>
<td>0.528</td>
<td>2.35</td>
<td>4.00 \cdot 10^{-4}</td>
<td>8.00 \cdot 10^{-3}</td>
<td>0</td>
</tr>
<tr>
<td>4</td>
<td>0.533</td>
<td>2.32</td>
<td>2.50 \cdot 10^{-4}</td>
<td>1.50 \cdot 10^{-2}</td>
<td>0</td>
</tr>
<tr>
<td>5</td>
<td>1.53</td>
<td>0.81</td>
<td>5.00 \cdot 10^{-4}</td>
<td>1.50 \cdot 10^{-2}</td>
<td>0</td>
</tr>
<tr>
<td><strong>Bi$_3$Fe$<em>5$O$</em>{12}$</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1</td>
<td>0.310</td>
<td>4.00</td>
<td>73.2</td>
<td>$&lt; 10^{-3}$</td>
<td>$&lt; 10^{-3}$</td>
</tr>
<tr>
<td>2</td>
<td>0.477</td>
<td>2.60</td>
<td>2.00</td>
<td>0.110</td>
<td>0.170</td>
</tr>
</tbody>
</table>
\[ N_{Er_2O_3}^2 = 1 + \frac{2.66}{1 - \left( \frac{0.150 \mu m}{\lambda} \right)^2 + i \cdot 3.61 \cdot 10^{-3} \cdot \left( \frac{0.150 \mu m}{\lambda} \right)} + \]
\[ \frac{1.07 \cdot 10^{-4}}{1 - \left( \frac{0.523 \mu m}{\lambda} \right)^2 + i \cdot 5.06 \cdot 10^{-3} \cdot \left( \frac{0.523 \mu m}{\lambda} \right)} + \]
\[ \frac{7.24 \cdot 10^{-5}}{1 - \left( \frac{0.528 \mu m}{\lambda} \right)^2 + i \cdot 6.81 \cdot 10^{-3} \cdot \left( \frac{0.528 \mu m}{\lambda} \right)} + \]
\[ \frac{4.63 \cdot 10^{-5}}{1 - \left( \frac{0.533 \mu m}{\lambda} \right)^2 + i \cdot 1.29 \cdot 10^{-2} \cdot \left( \frac{0.533 \mu m}{\lambda} \right)} + \]
\[ \frac{7.62 \cdot 10^{-4}}{1 - \left( \frac{1.53 \mu m}{\lambda} \right)^2 + i \cdot 3.70 \cdot 10^{-2} \left( \frac{1.53 \mu m}{\lambda} \right)}, \quad (6.7) \]

for Bi$_3$Fe$_5$O$_{12}$

\[ N_{Bi_3Fe_5O_{12}}^2 = 1 + \frac{4.58}{1 - \left( \frac{0.310 \mu m}{\lambda} \right)^2} + \]
\[ \frac{0.30}{1 - \left( \frac{0.490 \mu m}{\lambda} \right)^2 + i \cdot 0.09 \cdot \left( \frac{0.490 \mu m}{\lambda} \right)}. \quad (6.8) \]

Such formulae allow one to have an explicit view on the refractive indices and extinction coefficients of the materials. They are plotted in Figs 6.2 and 6.3 (alongside real and imaginary parts of $\varepsilon_{xy}$ of BIG).

There are special reasons for the introduction of several dipole transitions or, in other words, several terms in the interpolation formulae. For the titanium dioxide film, the short-wavelength transition is not enough to simulate the experimental transmittance well. An increase in $\Gamma$ leads to lowering the whole simulated transmittance. The long-wavelength transition with $\lambda_0 = 1.11 \mu m$ was therefore introduced to fit the transmittance adequately (see Figs 6.1a and 6.2a,b). Transmittance of the erbium oxide film may be fitted just by one transition with $\lambda_0 = 0.15 \mu m$. However, it is not possible to account for the resonance features around $0.53 \mu m$ and $1.53 \mu m$ (Fig. 6.1b). In order to take account of the resonance absorption, four additional dipole transitions were introduced with $\lambda_0$‘s = 0.523, 0.528, 0.533 and 1.53 $\mu m$ (see Figs 6.2a, b, c and d). The BIG transmittance (Fig. 6.1c) is not fitted by one short-wavelength dipole transition because of the absorption edge at around $0.55 \mu m$. The assumption of the second long-wavelength transition was made with the splitting of the excited state $\Delta \neq 0$. Such an assumption results in occurrence of the FR since $\varepsilon_{xy}$ is non-zero. The dipole transition with $\lambda_0 = 0.477 \mu m$ brings transmittance and FR to the desired values in the whole spectral range of our interest.
Figure 6.2. Refractive indices $n = \Re\{N\}$ and extinction coefficients $k = -\Im\{N\}$ of TiO$_2$ and Er$_2$O$_3$: a) refractive index and b) extinction coefficient of TiO$_2$ film as a function of wavelength; c) refractive index and d) extinction coefficient of Er$_2$O$_3$ film as a function of wavelength.

Eqs 2.33 and 2.34 in combination with $n$, $k$, $N$ and $N_\pm$ of the materials were used to simulate $\Theta_F$ and $T$. The simulations are depicted in Fig. 6.1 by red lines. It is apparent, the simulations by the Swanepoel and Višňovský formulae are in excellent agreement with the experimental results.

There is, however, a prominent discrepancy between the simulated and experimental spectra for Er$_2$O$_3$: the oscillation amplitude of the simulated spectrum is less than the one of the experimental spectrum. A slight shift of all the simulated transmittance spectra can also be noticed in the short-wavelength range. Elimination of both of them may be accomplished by performing certain improvements in the simulations to be discussed in Section 6.2.
Chapter 6. Simulations of Transmittance and Faraday Rotation

Figure 6.3. Bi$_3$Fe$_5$O$_{12}$ wavelength dependencies of: a) refractive index $n = \Re\{N\}$ (——), b) extinction coefficient $k = -\Im\{N\}$ (——), c) real part of the off-diagonal element of the permittivity tensor $\Re\{\varepsilon_{xy}\}$ (——), d) imaginary part of the off-diagonal element of the permittivity tensor $\Im\{\varepsilon_{xy}\}$ (——).

6.2 Photonic Crystals

540-nm (Er$_2$O$_3$/TiO$_2$)$_6$ Photonic Crystal

The 540-nm (Er$_2$O$_3$/TiO$_2$)$_6$ photonic crystal was deposited by PLD onto Corning glass 7059 substrate [26]. It was composed of two distributed Bragg reflectors (DBRs) (Er$_2$O$_3$/TiO$_2$)$_6$ and (TiO$_2$/Er$_2$O$_3$)$_6$ enclosing a cavity layer of Er$_2$O$_3$. The compositional shorthand may be written as (Er$_2$O$_3$/TiO$_2$)$_6$Er$_2$O$_3$2(TiO$_2$/Er$_2$O$_3$)$_6$. The layers in the DBRs had quarter-wave thicknesses $\lambda_{res}/4n_{TiO_2}/Er_2O_3(\lambda_{res}) = 53$ and 68.7 nm for TiO$_2$ and Er$_2$O$_3$, respectively. Here $\lambda_{res} = 0.54 \mu m$ denotes operational wavelength of the PC and $n_{TiO_2}/Er_2O_3(\lambda_{res})$ refractive index of TiO$_2$ or Er$_2$O$_3$ at $\lambda_{res}$. The cavity layer was of double thickness in comparison with an Er$_2$O$_3$ layer in a DBR (therefore, a power of 2 was added right after the cavity layer of Er$_2$O$_3$ in the compositional shorthand).
Whilst the PC is being grown, a slight deviation from the ideal layer thicknesses arises especially in the layer pairs after the cavity. In order to take this fact into account, a slight decrease of the layer thicknesses was introduced into the simulation (see Table 6.2).

### 980-nm \((\text{Bi}_3\text{Fe}_5\text{O}_{12}/\text{Sm}_3\text{Ga}_5\text{O}_{12})^5\) Magneto-Optical Photonic Crystal

The 980-nm \((\text{Bi}_3\text{Fe}_5\text{O}_{12}/\text{Sm}_3\text{Ga}_5\text{O}_{12})^5\) (BIG/SGG)\(^5\) magneto-optical photonic crystal was grown by RF magnetron sputtering \cite{27}. The MOPC was composed of the \([\text{Sm}_3\text{Ga}_5\text{O}_{12}/\text{Bi}_3\text{Fe}_5\text{O}_{12}]^5\) DBR having 5 layer pairs on the CMZGGG(111) substrate, cavity layer of BIG and the \([\text{Bi}_3\text{Fe}_5\text{O}_{12}/\text{Sm}_3\text{Ga}_5\text{O}_{12}]^5\) DBR having 5 layer pairs. The shorthand of the MOPC is written as \([\text{BIG/SGG}]^5\text{BIG}^2[\text{SGG/BIG}]^5\). The DBRs’ layers had quarter-wave thicknesses of \(\lambda_{res}/4n_{\text{BIG(SGG)}}(\lambda_{res}) = 96.3\) (125.9) nm, where \(n_{\text{BIG(SGG)}}(\lambda_{res})\) denotes refractive index of BIG (SGG) at \(\lambda_{res} = 0.98\) µm. The thickness of the cavity layer was 2 times larger than that of a BIG layer in the DBRs.

In the simulations, the refractive index of SGG was taken to equal the refractive index of CMZGGG in view of their close resemblance in optical properties.

### Simulation Results

Excellent agreement between the simulations and experimental data is demonstrated. All of the Fabry-Pérot oscillations are apparently reproduced by the simulations. The resonance features, including absorption edge in the case of the 980-nm MOPC, are simulated with a high extent of precision. The simulation routine is capable of copying even subtle details of the experimental transmittance and FR spectra. The aforementioned facts serve as unambiguous evidences of two circumstances: i) the layer thicknesses as well as optical properties of individual layers are reproduced well during the growth of the PCs, and ii) the simulation routine represents a stable means of predicting the optical and magneto-optical responses of photonic crystals and may be used to engineer multilayered structures with the desired transmittance and FR.

### Table 6.2

<table>
<thead>
<tr>
<th>Layer pair no.</th>
<th>Thicknesses, [nm]</th>
</tr>
</thead>
<tbody>
<tr>
<td>TiO(_2/\text{Er}_2\text{O}_3)</td>
<td></td>
</tr>
<tr>
<td>1-8</td>
<td>53/67.8</td>
</tr>
<tr>
<td>9-10</td>
<td>52/67.7</td>
</tr>
<tr>
<td>11-12</td>
<td>50.5/66.2</td>
</tr>
</tbody>
</table>
There are, however, several things to be pointed out. First, for the case of 980-nm MOPC, the resonance transmittance value of the simulated spectrum is in a very good agreement with the experimental one. Nevertheless, the simulated peak of FR is much lower in comparison with the experimental FR at resonance $\lambda_{res} = 0.98 \mu m$. Second, in regard to both the 540-nm PC and 980-nm MOPC, there is a slight shift of the Fabry-Pérot features of the simulated spectra towards longer wavelengths, especially in the short-wavelength range. Small differences in layer thicknesses and interdiffusion (if any) are easily discerned by short-wavelength light which might be the reason for that. The rationale for reduced FR has yet to be found.

The agreement between the simulation and experiment could be improved more by introduction of a larger number of dipole transitions. The problem, however, resides in two facts. Firstly, the standard nonlinear iteration algorithms cannot evaluate the whole set of the material parameters of TiO$_2$, Er$_2$O$_3$, and BIG when the number of dipole transitions is larger than two. The complexity of expressions requires a rather long time for such an evaluation which makes one to find the
6.2. Photonic Crystals

Figure 6.5. Transmittance and Faraday rotation spectra of the 980-nm
(Bi₃Fe₅O₁₂/Sm₃Ga₅O₁₂)⁶Bi₃Fe₅O₁₂²(Sm₃Ga₅O₁₂/Bi₃Fe₅O₁₂)⁶ photonic crystal: ○, experimental data and —, simulation results.

parameters by fitting the spectra with the naked eye. Secondly, in order to mitigate
the problem of finding reliable material parameters, one needs to obtain at least
some of them. The most trustworthy approach means extracting these parameters
from an absorption and FR, if any, spectra of the materials in bulk form. It is hardly
possible to grow TiO₂, Er₂O₃ and Bi₃Fe₅O₁₂ by either PLD or RF magnetron
sputtering in bulk form. This brings about the fact that absorption spectra of the
materials can be recorded using films only. The film absorption spectra, however,
are made up of Fabry-Pérot oscillations that do not allow accurate determination
of the resonance transition parameters.
Chapter 7

Conclusions

The thesis explored two types of photonic crystals \((\text{Er}_2\text{O}_3/\text{TiO}_2)^6\) and \((\text{BIG/}
\text{SGG})^5\) in the role of potential candidates for being optical amplifiers and optical isolators in PICs, respectively.

In order to achieve a suitable performance of PCs, the refractive index contrast between the constituent materials has to be the highest possible whereas absorption and interfacial roughness have to be the lowest. Therefore, tailoring of optical properties of \(\text{TiO}_2\) was performed to satisfy the requirements for the \((\text{Er}_2\text{O}_3/\text{TiO}_2)^6\) PC. This was done in terms of the three routines: deposition at elevated \(T_s\), regular annealing in thermodynamically equilibrium conditions and RTA. The first routine revealed suitable substrate temperatures of 300 and 400 °C to get high \(n\), low \(\alpha\) and \(\sigma\). The regular annealing showed the films satisfying the requirements are obtained after post-annealing at 300 °C. RTA revealed the suitability of film properties after rapid thermal annealing of the films at 500 °C. The highest \(n\) as well as lowest \(\alpha\) and \(\sigma\) is demonstrated by the film subjected to RTA at 500 °C.

Advanced modelling of transmittance for the \((\text{Er}_2\text{O}_3/\text{TiO}_2)^6\) and \((\text{BIG/}
\text{SGG})^5\) PCs and FR for the \((\text{BIG/} \text{SGG})^5\) was performed to establish a stable approach to engineering of PCs with the desired properties. The simulations for single film materials were done by the Swanepoel and Višňovský formulae. The aim was to extract wavelength dispersions of the diagonal and off-diagonal elements of the permittivity tensors of the materials. That enabled generation of dispersions of complex refractive indices for the materials. The combination of complex refractive indices with the \(4 \times 4\) matrix formalism resulted in the simulations of transmittance and Faraday rotation for the crystals. The modelling is found to give excellent forecast of transmittance and Faraday rotation for the \((\text{Er}_2\text{O}_3/\text{TiO}_2)^6\) and \((\text{BIG/}
\text{SGG})^5\) PCs.
Chapter 8

Papers Appended

The thesis presented is based on the four articles.

   My contribution: simulations of transmittance of single films and photonic crystals as well as extraction of wavelength dependences of refractive indices for the materials.

   My contribution: development of the approach to successful simulations of transmittance and Faraday rotation for the magneto-optical photonic crystals and its realisation.

   My contribution: AFM and XRD analyses, transmission spectroscopy, regular annealing and rapid thermal annealing treatments of the films as well as simulations of their transmittances.

   My contribution: being a short report of the previous paper, it has the same contribution of mine.
Bibliography


