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## **DOCTORAL THESIS**

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**Spectroscopic studies of new generation of optical and  
magneto-optical materials**

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Title: Spectroscopic studies of new generation of optical and magneto-optical materials

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Abstract:

In recent years, novel optical and magneto-optical devices have been proposed. This ranges from integrated photonic devices such as 3D holographic displays to magnetic recording, non-reciprocal photonic devices such as optical isolators and circulators or high-energy X-ray/gamma ray detectors. These devices, however, require suitable materials with tunable optical and magneto-optical properties. Presented thesis aims to systematically study such materials, namely  $Gd_xFe_{(100-x)}$ , magnetic garnets ( $Y_{3-x}Bi_xFe_5O_{12}$ ,  $Nd_2BiFe_{(5-x)}Ga_xO_{12}$ ,  $Nd_{0.5}Bi_{1.5}Fe_{(5-x)}Ga_xO_{12}$ ) and  $Ce_{(0.95-x)}Hf_xCo_{0.05}O_{(2-\delta)}$ . Systematic study is carried out by the combination of experimental methods of spectroscopic ellipsometry, magneto-optical Kerr effect spectroscopy and Faraday effect spectroscopy. Experimental data are confronted to theoretical calculations based on Yeh 4x4 matrix formalism. As a result, full permittivity tensor spectra of presented materials are derived and analyzed in terms of microscopic theory. This allows understanding and optimization of physical properties of studied materials which is important when increasing the application potential and suitability for variety of devices.

Keywords: Spectroscopy, Ellipsometry, Kerr effect, Faraday effect, Permittivity tensor

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# Introduction

Recent decades have been characteristic by a massive technology development that completely transformed our society. Technology has become smaller, faster, and more effective than ever. However, there is still a room for improvement, which has been demonstrated by multiple novel devices proposed by scientific community. In this category, one can find a huge variety ranging from 3D holographic displays to magnetic recording, integrated non-reciprocal photonic devices such as optical isolators and circulators, high-energy X-ray/gamma ray detectors and many more. This kind of novel technology, however, usually operates in nanoscale which complicates the situation. The reason is that nanoscale materials are not only difficult to prepare but their physical properties may also significantly differ from physical properties of their bulk forms. Therefore, the knowledge from the bulk material research can be used only to some extent. What is more, physical properties of materials in nanoscale are significantly influenced by surrounding materials (for example in a multilayer). This goes hand in hand with the compatibility requirement with the current technology (usually Si compatibility) which is particularly important. In addition, the huge variety of proposed highly specialized devices requires materials with tunable optical, magnetic, and magneto-optical (MO) properties. This type of tuning is usually performed by doping, composition or/and application of strain. These mechanisms must therefore be also understood and thoroughly researched.

To process all these inputs properly, one must come up with a parameter which characterizes optical and MO properties of studied materials completely. Moreover, this parameter must fully represent studied materials (together with the dimensions information) in any optical or MO calculation. This includes calculations of optical and MO response of complicated multilayered structures/nanostructures. Possibility to incorporate effect of the material on such a structure is especially important since structure represents proposed device. Therefore, the main advantage of such an approach would be possibility to design complicated optical and MO devices in the theoretical level before manufacturing any sample. As one can imagine, this can save a big amount of time, finances, and effort. The only parameter which meets all these



requirements is the permittivity tensor. Knowledge of the permittivity tensor spectra allows deep understanding of the optical and MO properties of the material. Furthermore, when combined with Yeh 4x4 matrix formalism, it allows calculations of interaction between electromagnetic radiation and multilayered structure.

For all these reasons combined, we devoted present work to the full permittivity tensor determination and analysis. This was done for three groups of promising novel materials:  $Gd_xFe_{(100-x)}$ , magnetic garnets ( $Y_{3-x}Bi_xFe_5O_{12}$ ,  $Nd_2BiFe_{(5-x)}Ga_xO_{12}$ ,  $Nd_{0.5}Bi_{2.5}Fe_{(5-x)}Ga_xO_{12}$ ) and  $Ce_{(0.95-x)}Hf_xCo_{0.05}O_{(2-\delta)}$ . To tune their physical properties properly, we investigated the influence of the material composition as well.

This thesis is organized as follows:

**Chapter 1** focuses on the understanding of the full permittivity tensor and its relation to the optical and MO properties of material. This includes energy absorption mechanisms as well as microscopic theory.

**Chapter 2** discusses some basic facts about the polarization state of optical waves and shows how MO parameters measured in experiment are related to the Jones matrix of the sample.

**Chapter 3** explains the wave equation in special geometries. Moreover, it explains general Yeh 4x4 matrix formalism for description of the optical and MO interactions in multilayers, necessary for theoretical calculations.

**Chapter 4** is devoted to the experimental techniques used for measurements of optical and MO spectra.

**Chapter 5** presents techniques used for the sample's preparation.

**Chapter 6** discusses obtained experimental and theoretical results. Based on the type of studied material, it is structured into three sub-chapters:  $Gd_xFe_{(100-x)}$ , magnetic garnets and  $Ce_{(0.95-x)}Hf_xCo_{0.05}O_{(2-\delta)}$ .

Finally, we devoted **Chapter 7** to the main conclusions.

# 1. PERMITTIVITY

Permittivity is a measure of proportionality that exists between electric displacement  $D$  and electric field  $E$  when forming an electric field in a medium. In simple terms, permittivity quantifies how an electromagnetic field affects, and is affected by a medium.

$$D = \varepsilon E \quad (1.1)$$

## 1.1. Complex permittivity function

If the medium is isotropic, the permittivity is a complex number. The reason behind this complexity is that response of the material to the external field depends on the field frequency  $\omega$ . This means that response is not instantaneous but casual. Therefore, we can represent this response as a phase difference. Complex numbers allow specification of phase and magnitude. Therefore, the permittivity becomes complex function defined by

$$D_0 e^{-i\omega t} = \varepsilon_1(\omega) E_0 e^{-i\omega t}. \quad (1.2)$$

Here  $E_0$  and  $D_0$  stand for the amplitude of electric displacement and the amplitude of electric field respectively;  $\varepsilon_1(\omega)$  is the complex permittivity function defined as

$$\varepsilon_1(\omega) = \varepsilon_{1r}(\omega) - i\varepsilon_{1i}(\omega). \quad (1.3)$$

Here,  $\varepsilon_{1r}$  refers to the real part that is related to the fraction of the energy dispersed by a medium. Consistently,  $\varepsilon_{1i}$  refers to the imaginary part that is related to the absorption loss (if it is positive) or gain (if it is negative). To summarize, the complex permittivity function represents optical properties of a material in terms of how material responds to the applied field. However, optical properties of the material can be also represented in terms of how the electromagnetic wave propagates in a material. For this purpose, we use the representation of refractive index  $n$  and absorption coefficient  $k$ . Refractive index is inversely proportional to the length of wave propagation in the material (length after which the phase of the wave changes by  $2\pi$ ). Similarly, absorption coefficient is inversely proportional to the distance in which the amplitude of the propagating wave decays to  $1/e$  of its original value. These constants are related by the equation

$$\varepsilon_{1r} - i\varepsilon_{1i} = (n - ik)^2. \quad (1.4)$$

The real and imaginary parts of the complex permittivity are not independent quantities, nor are the refractive index and absorption coefficient. These are connected by the Kramers-Kronig relations. These relations results from the requirement that material cannot respond to the applied field prior to its application.

$$n(\omega) = 1 + P \int_{-\infty}^{\infty} \frac{d\omega'}{\pi} \frac{k(\omega')}{\omega' - \omega}, \quad (1.5)$$

$$\varepsilon_{1r}(\omega) = 1 + P \int_{-\infty}^{\infty} \frac{d\omega'}{\pi} \frac{\varepsilon(\omega')\omega'}{\omega'^2 - \omega^2}. \quad (1.6)$$

We can derive complex permittivity and therefore all the optical functions of a material from spectroscopic ellipsometry (SE) measurements and analysis which will be discussed in more detail later in this work.

### 1.1.1. Energy absorption mechanisms

Types and strengths of optical absorption processes that occur in the material determine its optical properties and therefore its complex permittivity function completely. Material absorbs energy from a light beam by multiple mechanisms. The most important mechanisms are:

- a) **Interband absorption** refers to the case when an electron in a bound state in the material absorbs a single photon from the light beam and jumps to a higher energy level in the material.
- b) **Intraband absorption** refers to the case when an electron absorbs a photon from the light beam and jumps to a different energy state within the same band. This process usually requires photon emission or absorption. The only exception is when initial and final electron states occur at the same values of the crystal momentum.
- c) **Free carrier absorption** is a special case of the intraband absorption for conducting materials, which contain a gas of not bound, free carriers, which exhibit distinctive optical absorption.

### 1.1.2. Theoretical modeling of the complex permittivity function

As mentioned already, the shape and amplitude of the complex permittivity function of the material depends fully on types and strengths of optical absorption processes. To model these processes theoretically, scientists developed multiple theoretical models (usually referred as oscillators or terms), each of which is a function of light beam photon energy  $E$  (eV). In this part of the thesis, we discuss theoretical models used to model the complex permittivity function in this work.

#### a) Lorentz model

This model is based on classical theory, which describes an interaction of an optical wave with harmonically bound classical electron having a finite relaxation time. Classic version of Lorentz model is

$$\varepsilon_{1\_Lorentz} = \frac{AmpBrE_0}{E_0^2 - E^2 - i \cdot EBr} \quad (1.7)$$

Parameters  $E_0$ ,  $Amp$ ,  $Br$  denote the center energy, amplitude and the broadening parameter respectively [1, 2]. The shape that this model creates in the complex permittivity spectra can be seen in Figure 1.1.

#### b) Gaussian model

Gaussian model produces Gaussian line shape in  $\varepsilon_{li}$  with Kramers-Kronig consistent line shape for  $\varepsilon_{lr}$  [2, 3].

$$\varepsilon_{1\_Gaussian} = Amp \left\{ \begin{array}{l} \left( \Gamma \left( \frac{E - E_0}{\sigma} \right) + \Gamma \left( \frac{E + E_0}{\sigma} \right) \right) + \\ i \cdot \left( \exp \left[ - \left( \frac{E - E_0}{\sigma} \right)^2 \right] + \exp \left[ - \left( \frac{E + E_0}{\sigma} \right)^2 \right] \right) \end{array} \right\} \quad (1.8)$$

$$\sigma = \frac{Br}{2\sqrt{\ln(2)}} \quad (1.9)$$

Parameters  $E_0$ ,  $Amp$ ,  $Br$  and  $\sigma$  denote the center energy, amplitude, broadening and the conductance, respectively. The function  $\Gamma$  is a convergence series that produces a Kramers-Kronig consistent line shape for  $\varepsilon_{lr}$  [2, 3]. As one can see from Figure 1.1,

Gaussian model is particularly useful due to its ability to rapidly approach zero beyond the FWHM position.

### c) Tauc-Lorentz

This model reproduces the complex permittivity function of many amorphous materials particularly well. Tauc-Lorentz model (Figure 1.1) produces shape in  $\epsilon_{li}$  defined by equation

$$\epsilon_{li\_Tauc-Lorentz} = \left[ \frac{Amp E_0 Br (E - E_g)}{(E^2 - E_0^2)^2 + Br^2 E^2} \cdot \frac{1}{E} \right] \quad E > E_g \quad (1.10)$$

$$\epsilon_{li\_Tauc-Lorentz} = 0 \quad E \leq E_g. \quad (1.11)$$

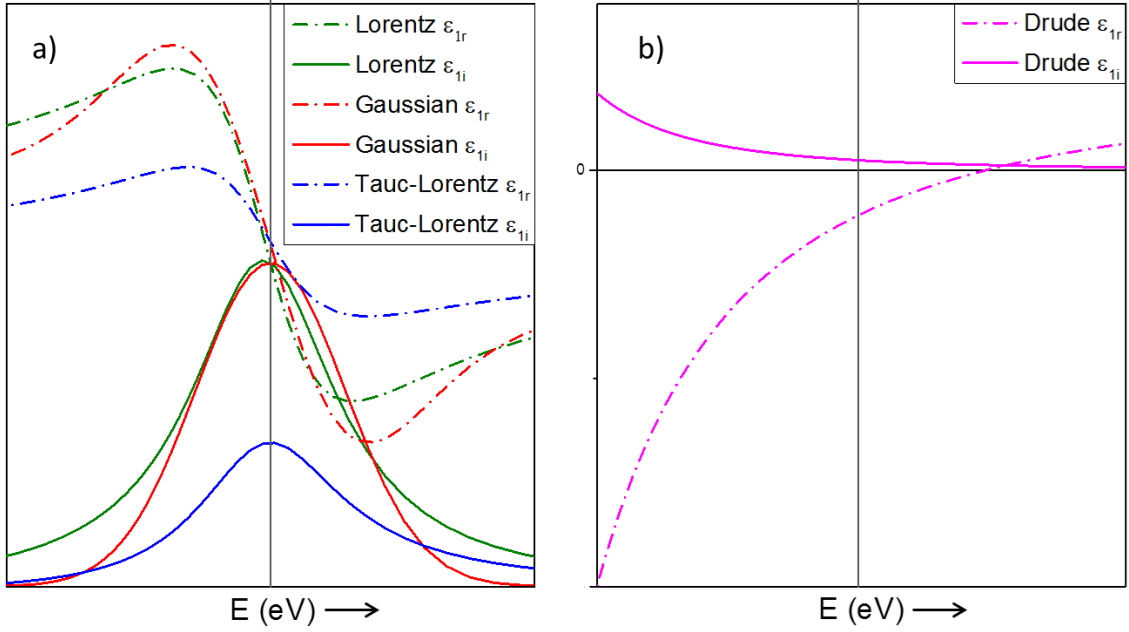
Parameters  $E_0$ ,  $Amp$ ,  $Br$ ,  $E_g$  denote the center energy, amplitude, broadening and bandgap energy, respectively. Function  $\epsilon_{lr}$  is produced using Kramers-Kronig relations [2, 4].

### d) Drude model

This model is a special case of Lorentz model where the center energy  $E_0$  equals zero. This model was developed to describe the free carrier effect on the complex permittivity function behavior. The model assumes that the microscopic behavior of free carriers in a solid may be treated with a gas of constantly moving carriers bouncing and re-bouncing off heavier static positive ions.

$$\epsilon_{l\_Drude} = \frac{-\hbar^2 q^2 N \mu}{\epsilon_0 (\mu m^* m_e E^2 + iq \hbar E)} \quad (1.12)$$

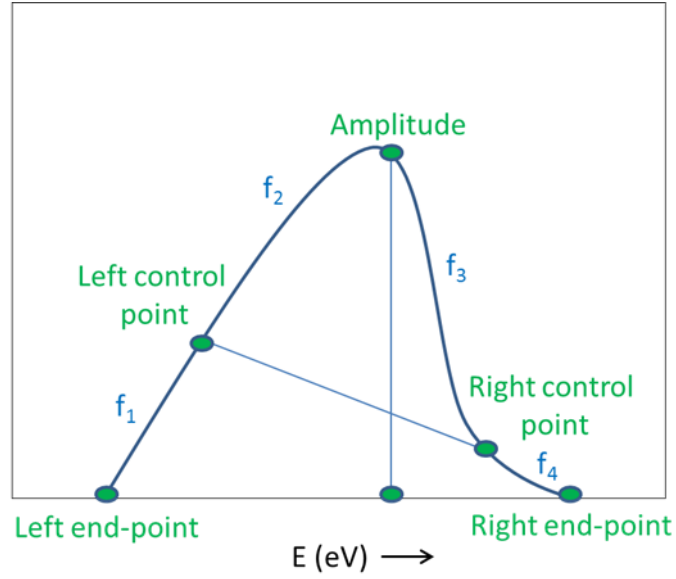
Parameters  $N$ ,  $\mu$ ,  $m^*$  denote the carrier concentration, carrier mobility and carrier effective mass, respectively. The physical constants are  $\hbar$  (Planck constant/2 $\pi$ ),  $q$  (electron charge),  $\epsilon_0$  (the vacuum dielectric constant) and  $m_e$  (the electron mass) [2, 5]. The shape this model creates in the complex permittivity spectra can be seen in Figure 1.1.



**Figure 1.1: The calculated spectra of  $\epsilon_{1r}$  and  $\epsilon_{1i}$  for a) Lorentz, Gaussian, and Tauc-Lorentz oscillators; b) Drude term.**

### e) Herzinger-Johs model

This model combines highly functional shape with Kramers-Kronig consistent properties, and it is useful especially when reproducing complicated complex permittivity function shapes of crystalline materials. The importance of this model is shape of the complex permittivity function it generates; however, its internal parameters have no direct physical meaning. This model consists of four polynomials spline functions  $f_1, f_2, f_3$  and  $f_4$  connected smoothly end-to-end. Functions  $f_1$  and  $f_3$  are equal zero at the endpoints. Variable fit parameters are  $E_0, Amp, Br, WL, WR, AL$  and  $AR$  that correspond to the center energy, amplitude, broadening, width of left side absorption region, width of right side absorption region, control point for left side and control point for right side respectively. The shape this model creates in  $\epsilon_{1i}$  spectra can be seen in Figure 1.2 [2, 6].



**Figure 1.2:** The calculated spectra of  $\epsilon_{li}$  for Herzinger-Johs model displaying polynomial spline functions  $f_1, f_2, f_3$  and  $f_4$ , as well as endpoints and control points.

## 1.2. Permittivity tensor

When we insert the isotropic material into the magnetic field, the field breaks the symmetry of the system. Therefore, we must treat the isotropic material in the magnetic field as anisotropic (all the materials investigated in this work exhibit anisotropy due to a magnetic ordering). Anisotropic system is generally characterized by a different direction of its electric field  $E$  and the electric induction  $D$  intensity vectors (1.1). Permittivity  $\epsilon$  in this system thus has a tensor character and we can express it as

$$\mathcal{E} = \begin{bmatrix} \mathcal{E}_{xx} & \mathcal{E}_{xy} & \mathcal{E}_{xz} \\ \mathcal{E}_{yx} & \mathcal{E}_{yy} & \mathcal{E}_{yz} \\ \mathcal{E}_{zx} & \mathcal{E}_{zy} & \mathcal{E}_{zz} \end{bmatrix}. \quad (1.13)$$

Since the field acts as a small perturbation of the isotropic material, we can express the permittivity tensor in the Cartesian representation as follows

$$\epsilon_{ij} = \epsilon_{ij}^0 + K_{ijk} M_k + G_{ijkl} M_k M_l. \quad (1.14)$$

Here,  $\epsilon_{ij}^0$  are components of the unperturbed permittivity,  $M_k$  are components of the magnetization vector;  $K_{ijk}$  and  $G_{ijkl}$  are the elements of the linear and quadratic MO

tensors responsible for linear and quadratic MO effects. In this work, we restrict ourselves to linear MO effects only. If we have magnetization parallel to the z-axis of the Cartesian coordinate system (the magnetic film-ambient interface is normal to the z-axis, light is propagating along the z-axis) we receive relations

$$\varepsilon_1 = \varepsilon_{xx} = \varepsilon_{yy} = \varepsilon_{zz}, \quad (1.15)$$

$$i\varepsilon_2 = -\varepsilon_{xy} = \varepsilon_{zx} \quad (1.16)$$

Permittivity tensor thus simplifies to the form

$$\begin{pmatrix} \varepsilon_1 & -i \cdot \varepsilon_2 & 0 \\ i \cdot \varepsilon_2 & \varepsilon_1 & 0 \\ 0 & 0 & \varepsilon_1 \end{pmatrix}. \quad (1.17)$$

All elements of the tensor have real and imaginary parts:

$$\varepsilon_1 = \varepsilon_{1r} - i \cdot \varepsilon_{1i}, \quad (1.18)$$

$$\varepsilon_2 = \varepsilon_{2r} - i \cdot \varepsilon_{2i}. \quad (1.19)$$

Off-diagonals  $\varepsilon_2$  are proportional to the magnetization in the sample. Therefore, if there is no magnetic field present, permittivity tensor reduces itself to a unit matrix of  $\varepsilon_1$ .

### 1.2.1. Microscopic theory

Microscopic theory relates full permittivity tensor spectra to energy level splitting and transition probabilities. As mentioned earlier, when there is a magnetic field applied, off-diagonals of the permittivity tensor  $\varepsilon_2$  have finite values. From microscopic point of view, there are three distinct mechanisms producing these finite values:

- 1) An unequal population of states related to the *spin polarization of the ground state* displays the opposite contributions to  $\varepsilon_2$ .
- 2) Variations in the energy differences between two states caused by:
  - *Zeeman splitting* of the energy levels when the external field acts on the orbital electronic motion
  - *Spin-orbit splitting*



- 3) Perturbations caused by **spin orbit coupling** effect on wave functions of occupied or unoccupied states or on the kinetic momentum operator.

To summarize, finite values of the  $\varepsilon_2$  mean that there are new types of optical transitions in the material, MO transitions, which exist only when the magnetic field is applied. In this work, we will consider two types of these transitions: Dia and Para transitions.

**a) Dia transitions** refer to spin and electric-dipole allowed transitions between an orbital singlet ground state and an excited state split by the combined effect of exchange field and spin-orbit coupling [7, 8]. These transitions can be described by an oscillator term, in which  $\varepsilon_2$  behaves as:

$$\varepsilon_2 = \frac{\omega_p^2 f \Delta L}{2\omega_0} \frac{(\omega_0 - \omega)^2 - \Gamma_0^2 + 2i\Gamma_0(\omega_0 - \omega)}{[(\omega_0 - \omega)^2 + \Gamma_0^2]^2} \quad (1.20)$$

Here  $\Delta$ ,  $\omega_0$ ,  $\Gamma_0$  and  $f$  are the separation between the levels caused by spin-orbit coupling, center frequency, half width at half-height of the transition and the oscillator strength, respectively.  $L$  is the Lorentz-Lorentz local field correction defined as  $[(n^2+2)/3]^2$ , where  $n$  is the refraction index. Dia transition behavior in  $\varepsilon_2$  close to the center frequency  $\omega_0$  is schematically shown in Figure 1.3(a). We can observe a bell-shaped behavior for the real part  $\varepsilon_{2r}$  and dispersive behavior for the imaginary part  $\varepsilon_{2i}$ . At center frequency  $\omega=\omega_0$ ,  $\varepsilon_{2i}=0$ ;  $\varepsilon_{2r}$  has a maximum value

$$(\varepsilon_{2r})_{\max} = \frac{\omega_p^2 f \Delta L}{2\omega_0 \Gamma_0^2} \quad (1.21)$$

and  $\varepsilon_{1i}$  shows a resonant behavior with maximum value of  $\varepsilon_{1i}$

$$(\varepsilon_{1i})_{\max} = \frac{\omega_p^2 f L}{2\omega_0 \Gamma_0} \quad (1.22)$$

**b) Para transitions**

In the case of Para transitions neither the ground state nor the final state is split. However, the oscillator strengths for right circularly polarized light  $f_+$  and left

circularly polarized light  $f$  are different [7, 8]. These transitions can be described by an oscillator term, which in  $\epsilon_2$  behaves as:

$$\epsilon_2 = \frac{\omega_p^2 f df L}{2} \frac{\omega(\omega_0^2 - \omega^2 - \Gamma_0^2) - i\Gamma_0(\omega_0^2 + \omega^2 + \Gamma_0^2)}{\omega_0 [(\omega_0^2 + \omega^2 + \Gamma_0^2)^2 + 4\omega^2 \Gamma_0^2]^2}. \quad (1.23)$$

Here  $df$  is the fractional dichroism defined as

$$df = \frac{f_- - f_+}{f_- + f_+}. \quad (1.24)$$

Para transition behavior in  $\epsilon_2$  close to the center frequency  $\omega_0$  is schematically shown in Figure 1.3(b). In this case, we can observe dispersive behavior for  $\epsilon_{2r}$  and dissipative behavior for  $\epsilon_{2i}$ . At  $\omega = \omega_0$ ,  $\epsilon_{2r} = 0$ ;  $\epsilon_{2i}$  has a maximum value

$$(\epsilon_{2i})_{\max} = \frac{\omega_p^2 f df L}{\omega_0} = df (\epsilon_{1i})_{\max}. \quad (1.25)$$

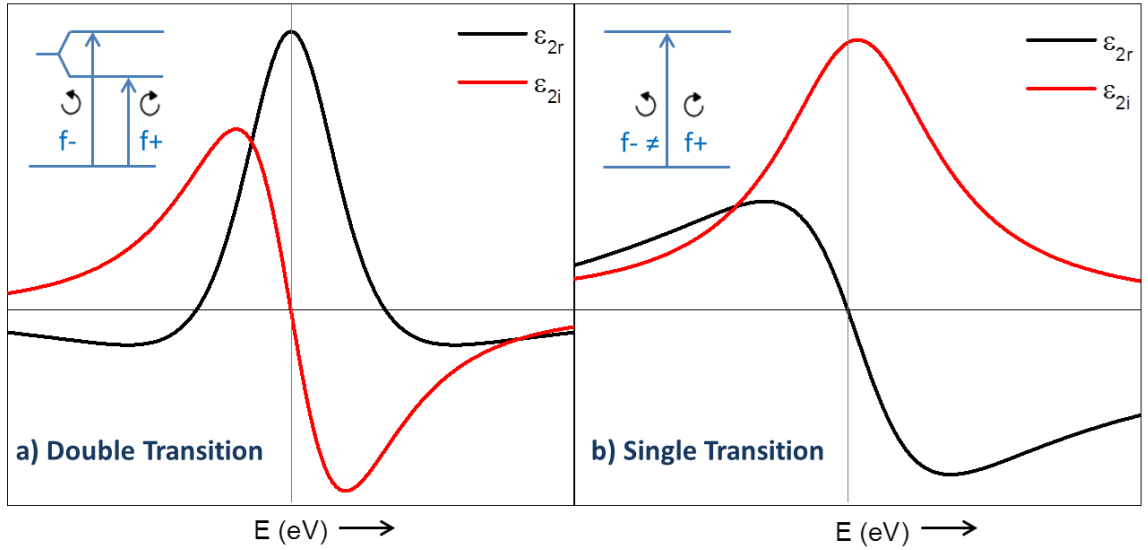


Figure 1.3: The calculated spectra of  $\epsilon_{2r}$  and  $\epsilon_{2i}$  for a) Dia transition and b) Para transition.

## 2. LIGHT POLARIZATION, OPTICAL AND MAGNETOOPTICAL EFFECTS

This chapter is a brief introduction to the light polarization and Jones vector formalism, which is a powerful tool when calculating polarization properties of light. Furthermore, we discussed in here SE and MO effects which are all based on the change in the polarization state of light upon reflection or transmission.

### 2.1. Light polarization

Light is understood as a general transverse electromagnetic radiation. Polarization of an electro-magnetic light wave is given by the time-dependent evolution of the electric field vector  $E$ . There are many ways to describe the polarization state of fully polarized electromagnetic waves. Since the electromagnetic light wave is generally elliptically polarized, we most often operate with the parameters of the polarization ellipse (Figure 2.1): azimuth  $\theta$ , ellipticity  $e$ ; and Jones vector formalism [9, 10].

- Azimuth,  $(-\pi/2 \leq \theta < \pi/2)$  is an oriented angle between the x-axis of the Cartesian coordinate system and the semi-major axis of the polarization ellipse. In this work, we choose the positive sign of the azimuth for the counterclockwise orientation.
- Ellipticity  $e$ ,  $(-1 \leq e \leq 1)$  is a proportion of minor  $a$  and  $b$  axes of the polarization ellipse. At the same time, we introduce variable ellipticity (ellipticity angle)  $\epsilon$ . In this work, we choose the positive sign of the ellipticity for the clockwise orientation.

$$e = \pm \frac{a}{b} = \tan \epsilon \quad (2.1)$$

The polarization state of light waves can be completely determined by using parameters of the polarization ellipse.

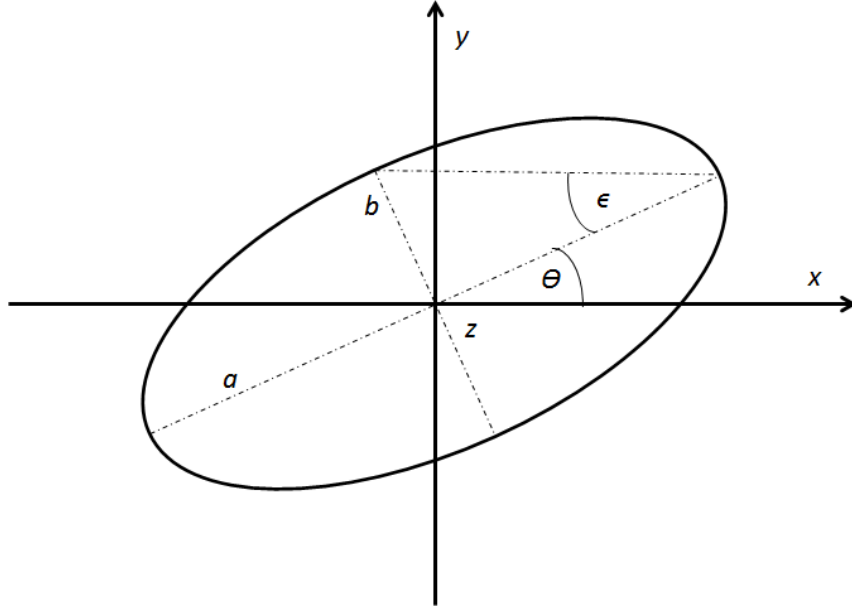


Figure 2.1: The polarization ellipse and basic MO parameters.

## 2.2. Jones vector formalism

Jones vector formalism describes the polarization state of light by two-dimensional complex vector whose coordinates are given by the choice of the polarization base. If we choose the polarization base as two orthogonal polarization amplitudes  $a_1$  and  $a_2$ , with a phase difference  $\delta$ , and if we define an angle  $\alpha$  as  $\tan \alpha = a_2/a_1$ , then we can express normalized Jones vector in a form

$$J = \begin{bmatrix} \cos \alpha \\ \sin \alpha e^{i\delta} \end{bmatrix}. \quad (2.2)$$

In the Cartesian-base of linear polarizations, we can express Jones vectors for linear and general elliptical polarizations as follows:

- linear polarization along the  $x$  and  $y$  axis [10]

$$E_x = \begin{bmatrix} 1 \\ 0 \end{bmatrix}, E_y = \begin{bmatrix} 0 \\ 1 \end{bmatrix}, \quad (2.3)$$

- general elliptical polarization [10]

$$J_{xy} = \begin{bmatrix} \cos \theta \cos \epsilon - i \sin \theta \sin \epsilon \\ \sin \theta \cos \epsilon + i \cos \theta \sin \epsilon \end{bmatrix}. \quad (2.4)$$

General complex Jones vector describes the polarization ellipse well, but it contains information of the initial phase and amplitude. However, this information is redundant for the description of the polarization state of light. For this reason, we introduce a complex number, complex polarization parameter  $\chi$ , defined as a proportion of the first and second component of the Jones vector. Complex polarization parameter for the polarization ellipse expressed by the Jones vector (2.4) thus has following form [9]

$$\chi_{xy} = \frac{E_y}{E_x} = \frac{\sin \theta \cos \epsilon + i \cos \theta \sin \epsilon}{\cos \theta \cos \epsilon - i \sin \theta \sin \epsilon} = \frac{\tan \epsilon \theta + i \tan \epsilon}{1 - i \tan \theta \tan \epsilon}. \quad (2.5)$$

For small angles  $\theta$  and  $\epsilon$ , and after approximations  $\tan \theta = \theta$  and  $\tan \epsilon = \epsilon$  (restricted to members of the first order), we can rewrite this expression as

$$\chi_{xy} \approx \theta + i\epsilon. \quad (2.6)$$

Jones formalism in Cartesian representation enables simplified and effective description of the polarized light properties after reflection or transmission on the sample. In here we choose the base of Cartesian system defined by  $s$  ( $E$  in the plane of incidence) and  $p$  ( $E$  perpendicular to the plane of incidence) polarizations. In this base, it is possible to express the effect of sample on the polarization state of incident light beam upon reflection by a reflection matrix  $S_R$ . Similarly, we can express the effect of sample on the polarization state of incident light beam upon transmission by a transmission matrix  $S_T$  [9].

$$S_R = \begin{bmatrix} r_{ss} & r_{sp} \\ r_{ps} & r_{pp} \end{bmatrix}, \quad (2.7)$$

$$S_T = \begin{bmatrix} t_{ss} & t_{sp} \\ t_{ps} & t_{pp} \end{bmatrix}. \quad (2.8)$$

In the first approximation, diagonal elements of  $S_R$  and  $S_T$  matrices correspond to Fresnel coefficients. On the other hand, off-diagonal elements (in the first approximation linear in magnetization) describe the interaction between  $s$  and  $p$  waves that is occurring in an anisotropic media.

Let us describe the incident wave by Jones vector  $J_{in}$ ; reflected wave by Jones vector  $J_{out}^R$  and transmitted wave by Jones vector  $J_{out}^T$ . Using reflection and transmission matrices, we can define relations between these as follows [11]

$$J_{out}^R = S_R \cdot J_{in}, \quad J_{out}^T = S_T \cdot J_{in}. \quad (2.9)$$

### 2.3. Spectroscopic Ellipsometry

Spectroscopic ellipsometry is a very sensitive measurement technique that uses polarized light to characterize thin films, surfaces and material microstructures [12]. Ellipsometry is an useful technique which allows to determine material properties such as: film thickness, refractive index, complex permittivity function, conductance, absorption, surface roughness, interfacial regions, sample composition, film composition, crystallinity, optical anisotropy, uniformity, alloy ratio and depth profile of material properties. This technique measures the change in the polarization state of the reflected or transmitted light and compares it to a model (Figure 2.2).

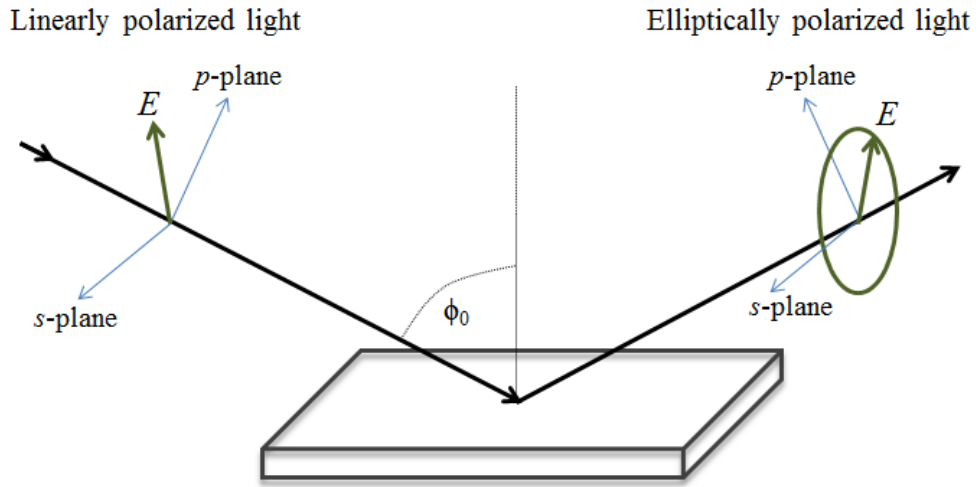


Figure 2.2: Geometry of an ellipsometry experiment

It is possible to derive diagonal elements of the permittivity tensor from SE data analysis. We can express the change in the polarization state of a reflected beam by SE parameters: amplitude ratio  $\psi$  and phase difference  $\Delta$ . These parameters are defined as

$$\tan \psi \cdot e^{i\Delta} = \rho = \frac{r_{pp}}{r_{ss}}. \quad (2.10)$$

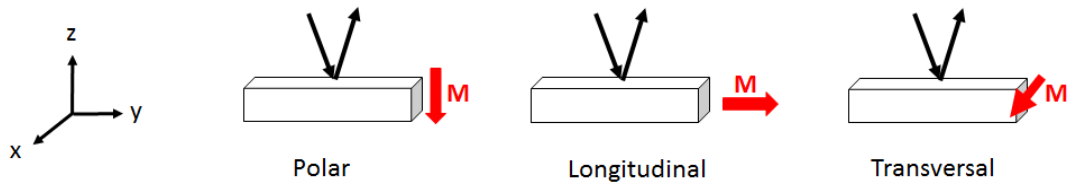
In this equation,  $\tan\psi$  is the magnitude of the reflectivity ratio and  $\Delta$  is the phase change between  $s$  and  $p$  polarized light. The  $r_{pp}$  and  $r_{ss}$  are amplitude reflection coefficients for  $s$  and  $p$  polarization measured from the alternating current signal [2]. The important part of SE analysis is the proper parametrization of the dispersion of unknown optical functions. For purposes of parameterization we use oscillators/terms defined in the subchapter 1.1.2 fitted to resulted optical functions spectra.

## 2.4. Magneto-optical effects

In this work, we use MOKE and MO Faraday effect to study the physical properties of magnetic materials and magnetic layered structures. These phenomena are characteristic by changes in the polarization state of the reflected or transmitted light caused by magnetic ordering [9]. In the MOKE experiment, we study these changes upon light reflection. Contrarily, in the MO Faraday effect experiment we study these changes upon light transmission.

### a) Magneto-optical Kerr effect

We can categorize the MOKE based on: the mutual orientation of the incidence plane, the reflection plane of the sample and the magnetization vector  $M$ . Based on these parameters, we recognize three basic configurations: polar, longitudinal and transversal (Figure 2.3).



**Figure 2.3: Basic configurations for measuring the MOKE.**

Considering the geometry of these configurations, we can simplify the permittivity tensor to forms

$$\epsilon_p = \begin{bmatrix} \epsilon_1 & -i\epsilon_2 & 0 \\ i\epsilon_2 & \epsilon_1 & 0 \\ 0 & 0 & \epsilon_1 \end{bmatrix}, \quad (2.11)$$

$$\varepsilon_L = \begin{bmatrix} \varepsilon_1 & 0 & -i\varepsilon_2 \\ 0 & \varepsilon_1 & 0 \\ i\varepsilon_2 & 0 & \varepsilon_1 \end{bmatrix}, \quad (2.12)$$

$$\varepsilon_T = \begin{bmatrix} \varepsilon_1 & 0 & 0 \\ 0 & \varepsilon_1 & -i\varepsilon_2 \\ i\varepsilon_2 & 0 & \varepsilon_1 \end{bmatrix}. \quad (2.13)$$

where  $\varepsilon_P$  stands for permittivity tensor in the polar,  $\varepsilon_L$  in the longitudinal and  $\varepsilon_T$  in the transversal configuration. In these geometries, also reflection matrix  $S_R$  defined by equation (2.7) takes different forms

$$S_{R\_polar} = \begin{bmatrix} r_{ss} & r_{sp} \\ r_{ps} & r_{pp} \end{bmatrix}, \quad S_{R\_long} = \begin{bmatrix} r_{ss} & -r_{ps} \\ r_{ps} & r_{pp} \end{bmatrix}, \quad S_{R\_trans} = \begin{bmatrix} r_{ss} & 0 \\ 0 & r_{pp} \end{bmatrix}. \quad (2.14)$$

Physical meaning of  $S_R$  matrix elements then allows defining of MO parameters, Kerr rotation  $\theta_K$  and Kerr ellipticity  $\epsilon_K$ , for  $s$  and  $p$  polarized waves as follows [9]

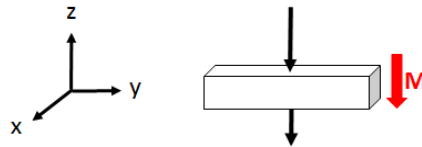
$$\theta_{Ks} - i\delta_{Ks} \approx \Phi_{Ks} \equiv -\frac{r_{ps}}{r_{ss}}, \quad (2.15)$$

$$\theta_{Kp} - i\delta_{Kp} \approx \Phi_{Kp} \equiv \frac{r_{sp}}{r_{pp}}. \quad (2.16)$$

In case of normal incidence of the light beam, given sign convention provides equal values of MO parameters  $\Phi_{Ks} = \Phi_{Kp}$ .

## b) Magneto-optical Faraday effect

We usually measure the MO Faraday effect in the configuration shown in the Figure 2.4.



**Figure 2.4: Basic configuration for measuring the Faraday effect.**

Considering the geometry of this configuration, we can simplify the permittivity tensor to the form



$$\boldsymbol{\varepsilon}_F = \begin{bmatrix} \varepsilon_1 & -i\varepsilon_2 & 0 \\ i\varepsilon_2 & \varepsilon_1 & 0 \\ 0 & 0 & \varepsilon_1 \end{bmatrix}. \quad (2.17)$$

In this geometry are the transmission matrix  $S_T$  elements defined by the equation (2.8). Therefore, we can define MO parameters, Faraday rotation  $\theta_F$  and Faraday ellipticity  $\epsilon_F$ , for  $s$  and  $p$  polarized waves as follows [9]

$$\theta_{Fs} - i\epsilon_{Fs} \approx \Phi_{Fs} \equiv -\frac{t_{ps}}{t_{ss}}, \quad (2.18)$$

$$\theta_{Fp} - i\epsilon_{Fp} \approx \Phi_{Fp} \equiv -\frac{t_{sp}}{t_{pp}}. \quad (2.19)$$

In case of normal incidence of the light beam, given sign convention provides equal values of MO parameters  $\Phi_{Fs} = \Phi_{Fp}$ .

# 3. ELECTROMAGNETIC WAVES IN ANISOTROPIC MEDIUM

In the previous chapter, we briefly introduced the description of the polarization state of light, SE and MO effects. However, for a deeper analysis of the optical and MO response of thin films and multilayers, we need a macroscopic analysis of the optical interaction of the polarized light with a substance. We will discuss this interaction in following paragraphs devoted to the behavior of polarized light in anisotropic media.

## 3.1. Wave equation in anisotropic media

We can describe the polarized monochromatic electromagnetic plane wave incident from the vacuum on the anisotropic environment without free charges by Maxwell's equations [9, 10]

$$\nabla \times E + \frac{\partial B}{\partial t} = 0, \quad (3.1)$$

$$\nabla \cdot D = 0, \quad (3.2)$$

$$\nabla \times H - \frac{\partial D}{\partial t} = 0, \quad (3.3)$$

$$\nabla \cdot B = 0, \quad (3.4)$$

where material equations are

$$D = \varepsilon_0 \varepsilon E, \quad (3.5)$$

$$B = \mu_0 \mu H. \quad (3.6)$$

From Maxwell's equations, we derive the wave equation in the traditional way

$$\Delta E - \frac{\varepsilon}{c^2} \frac{\partial^2 E}{\partial t^2} - \nabla(\nabla \cdot E) = 0. \quad (3.7)$$

We are looking for its solution in the shape of a plane wave

$$E = E_0 e^{i(\omega t - k \cdot r)}. \quad (3.8)$$

If we introduce a reduced wave vector  $N$

$$N = \frac{c}{\omega} k, \quad (3.9)$$

we may rewrite the original wave equation into a matrix form [9]

$$\begin{bmatrix} \epsilon_{xx} - N_y^2 - N_z^2 & \epsilon_{xy} & \epsilon_{xz} \\ \epsilon_{yx} & \epsilon_{yy} - N_z^2 & \epsilon_{yz} - N_y N_z \\ \epsilon_{zx} & \epsilon_{zy} - N_y N_z & \epsilon_{zz} - N_y^2 \end{bmatrix} \begin{bmatrix} E_{0x} \\ E_{0y} \\ E_{0z} \end{bmatrix} = 0. \quad (3.10)$$

In this derivation we assumed the plane of incidence perpendicular to the x-axis ( $N_x = 0$ ), and thus the component of the reduced wave vector  $N_y$  (derived from a Snell's law) in shape

$$N_y = N_0 \sin \varphi_0, \quad (3.11)$$

Here,  $N_0$  is a real refractive index of the isotropic medium and  $\varphi_0$  is an angle of incidence of the electromagnetic wave. For the case of zero determinant, we can find a nontrivial solution of the equation (3.10) as a characteristic equation of 4-th order for  $N_z$

$$\begin{aligned} & N_z^4 \epsilon_{zz} + N_z^3 [N_y (\epsilon_{yz} + \epsilon_{zy})] - \\ & - N_z^2 [\epsilon_{yy} (\epsilon_{zz} - N_y^2) + \epsilon_{zz} (\epsilon_{xx} - N_y^2) - \epsilon_{xz} \epsilon_{zx} - \epsilon_{yz} \epsilon_{zy}] - \\ & - N_y N_z [(\epsilon_{xx} - N_y^2) (\epsilon_{yz} + \epsilon_{zy}) - \epsilon_{xy} \epsilon_{zx} - \epsilon_{yx} \epsilon_{xz}] + \\ & + \epsilon_{yy} [(\epsilon_{xx} - N_y^2) (\epsilon_{zz} - N_y^2) - \epsilon_{xz} \epsilon_{zx}] - \\ & - \epsilon_{xy} \epsilon_{yz} (\epsilon_{zz} - N_y^2) - \epsilon_{yz} \epsilon_{zy} (\epsilon_{xx} - N_y^2) + \epsilon_{xy} \epsilon_{zx} \epsilon_{yz} + \epsilon_{yx} \epsilon_{xz} \epsilon_{zy} = 0. \end{aligned} \quad (3.12)$$

The roots of the characteristic equation (3.12) correspond to four proper modes of light propagation in anisotropic media  $N_{zj}$ . Two of these modes propagate with  $+k$  in the forward direction and two with  $-k$  in the reverse direction. Eigenvectors of these modes for the general permittivity tensor are

$$e_j = \begin{bmatrix} -\epsilon_{xy} (\epsilon_{zz} - N_y^2) + \epsilon_{xz} (\epsilon_{zy} + N_y N_{zj}) \\ (\epsilon_{zz} - N_y^2) (\epsilon_{xx} - N_y^2 - N_{zj}^2) - \epsilon_{xz} \epsilon_{zx} \\ -(\epsilon_{xx} - N_y^2 - N_{zj}^2) (\epsilon_{zy} - N_y N_{zj}) - \epsilon_{zx} \epsilon_{xy} \end{bmatrix}. \quad (3.13)$$

Linear superposition of these four modes of light propagation in the anisotropic media gives a general solution of the wave equation

$$E = \sum_{j=1}^4 E_{0j} e_j e^{i[\omega t - \frac{\omega}{c}(N_y y + N_z z)]}. \quad (3.14)$$

We can greatly simplify this solution when working with the general anisotropic medium in polar or longitudinal configuration.

- In the polar configuration, we define the permittivity tensor by equation (2.11). In this case, wave equation leads to a simplified characteristic equation whose solutions correspond to the four proper modes of the light propagation  $N_{zj}$  [9]

$$N_{z1} = N_{z+}, \quad N_{z2} = -N_{z+}, \quad N_{z3} = N_{z-}, \quad N_{z4} = -N_{z-}, \quad (3.15)$$

where

$$N_{z\pm} = \sqrt{(\varepsilon_1 - N_y^2) \left(1 \pm \frac{\varepsilon_2}{\sqrt{\varepsilon_1(\varepsilon_1 - N_y^2)}}\right)}. \quad (3.16)$$

- In the longitudinal configuration, we define the permittivity tensor by equation (2.12). In this case, wave equation leads to the characteristic equation with solutions  $N_{zj}$  [9]

$$N_{z1} = N_{z+}, \quad N_{z2} = -N_{z+}, \quad N_{z3} = N_{z-}, \quad N_{z4} = -N_{z-}, \quad (3.17)$$

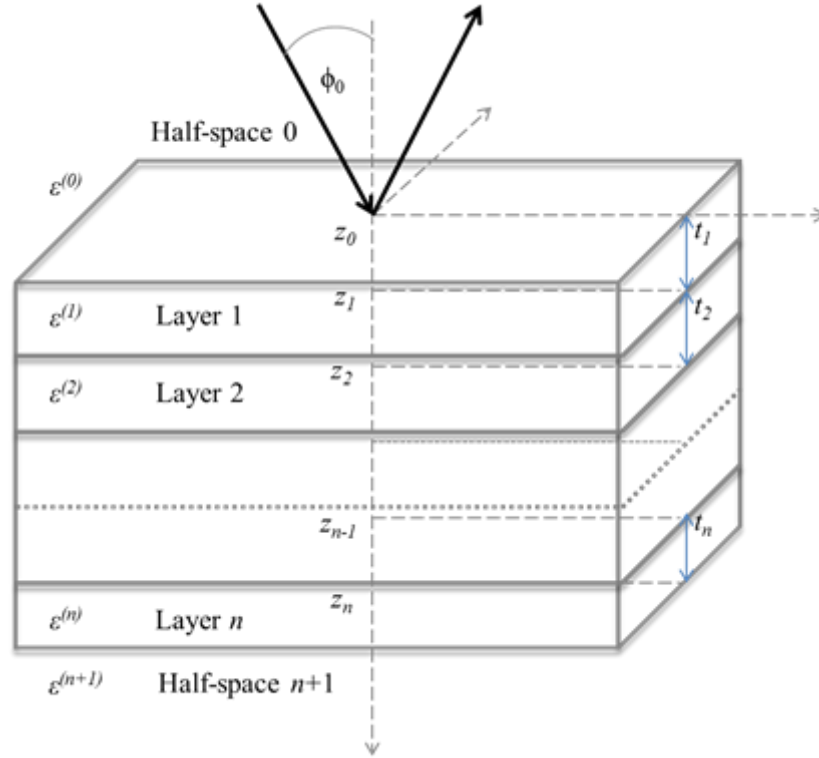
where

$$N_{z\pm} = \sqrt{\varepsilon_1 - N_y^2 - \frac{\varepsilon_2}{2\varepsilon_1} (\pm \sqrt{(4\varepsilon_1 N_y^2 + \varepsilon_2^2 - \varepsilon_2)}}. \quad (3.18)$$

### 3.2. Yeh 4x4 matrix formalism

Up to this point we have covered only the propagation of electromagnetic waves in anisotropic environment. However, for the evaluation of the MO experiments on multilayers, we must extend this description. The reason is that the interaction of light in a layered structure with sufficiently thin layers depends on all the layers contribution. This fact forces us to introduce a formalism that allows clear

description of the interaction of electromagnetic radiation with magnetic multilayer, Yeh 4x4 matrix formalism [13].



**Figure 3.1: Anisotropic multilayered structure. The individual layers are characterized by electric permittivity  $\epsilon^{(n)}$  and the thickness  $t^{(n)}$ ,  $n = 1, 2, \dots, n$ . Angle of incidence in the half-space 0 is labeled  $\phi_0$ . Planar interfaces of layers are perpendicular to the  $z$ -axis, plane of incidence is perpendicular to the  $x$ -axis.**

Let us consider multilayer made of  $n$  layers with mutually parallel interfaces perpendicular to the  $z$ -axis (Figure 3.1). Neighboring isotropic half-spaces 0 and  $n + 1$  will be described by scalar electric permittivity  $\epsilon(0)$  and  $\epsilon(n + 1)$ . Boundary conditions of continuity of vectors  $E$  and  $B$  tangential components on individual interfaces allow us to tie relations of electromagnetic fields in  $n$  and  $n + 1$  layers. It can be done in the representation of linear transformations of proper modes, which can be expressed in matrix form as

$$D^{(n-1)} E_0^{(n-1)}(z_n - 1) = D^{(n)} P^{(n)} E_0^{(n)}(z_n). \quad (3.19)$$

Here P stands for a Propagation matrix

$$P_{ij}^{(n)} = \delta_{ij} \exp(i \frac{\omega}{c} \bar{N}_{zj}^{(n)} t_n), \quad (3.20)$$

where  $\delta_{ij}$  is a Kronecker delta and  $t_n$  represents a thickness of the  $n$ -th layer.  $D$  represents a Dynamical matrix whose elements have form [9, 13, 14]

$$D_{1j}^{(n)} = -\varepsilon_{xy}^{(n)}(\varepsilon_{zz}^{(n)} - N_y^2) + \varepsilon_{xz}^{(n)}(\varepsilon_{zy}^{(n)} - N_y N_{zj}^{(n)}), \quad (3.21)$$

$$D_{2j}^{(n)} = N_{zj}^{(n)}[-\varepsilon_{xy}^{(n)}(\varepsilon_{zz}^{(n)} - N_y^2) + \varepsilon_{xz}^{(n)}(\varepsilon_{zy}^{(n)} - N_y N_{zj}^{(n)})], \quad (3.22)$$

$$D_{3j}^{(n)} = (\varepsilon_{zz}^{(n)} - N_y^2)(\varepsilon_{xx}^{(n)} - N_y^2 - N_{zj}^{(n)2}) - \varepsilon_{xz}^{(n)}\varepsilon_{zx}^{(n)}, \quad (3.23)$$

$$D_{4j}^{(n)} = -(\varepsilon_{xx}^{(n)} - N_y^2 - N_{zj}^{(n)2})(N_y \varepsilon_{zy}^{(n)} + N_{zj} \varepsilon_{zz}^{(n)}) + N_{zj}^{(n)} \varepsilon_{xz}^{(n)} \varepsilon_{zx}^{(n)} + N_y \varepsilon_{zx}^{(n)} \varepsilon_{xy}^{(n)}. \quad (3.24)$$

By modification of equation (3.19) it is possible to introduce the transfer matrix  $T$ , which ties up the field between  $(n-1)$  and the  $n$ -th layer.

$$E_0^{(n-1)}(z_{n-1}) = (D^{(n-1)})^{-1} D^{(n)} P^{(n)} E_0^{(n)}(z_n) = T^{(n-1,n)} E_0^{(n)}(z_n). \quad (3.25)$$

By application of the equation (3.25) for each interface of the multilayer, it is possible to express relationship between the incident waves passing through the first interface and the wave passing through the  $n$ -th interface as

$$E_0^{(0)}(z_0) = \left( \prod_{n=1}^{n+1} T^{(n-1,n)} \right) E_0^{(n+1)}(z_n) = M E_0^{(n+1)}(z_n). \quad (3.26)$$

Relation (3.26) defines a matrix of the multilayered structure  $M$ . This matrix can characterize any layered anisotropic structure, and it is sufficient to determine reflection coefficients of the structure. If we consider that the source of electromagnetic radiation is strictly in the upper half-space  $0$  (from lower half-space is not coming any radiation and so  $E_{02}$  and  $E_{04}$  are equal zero), we can express reflection coefficients by components of matrix  $M$  as [9, 13]

$$r_{12} = \left( \frac{E_{02}^{(0)}(z_0)}{E_{01}^{(0)}(z_0)} \right)_{E_{03}^{(0)}(z_0)=0} = \frac{M_{21}M_{33} - M_{23}M_{31}}{M_{11}M_{33} - M_{13}M_{31}}, \quad (3.27)$$

$$r_{14} = \left( \frac{E_{04}^{(0)}(z_0)}{E_{01}^{(0)}(z_0)} \right)_{E_{03}^{(0)}(z_0)=0} = \frac{M_{41}M_{33} - M_{43}M_{31}}{M_{11}M_{33} - M_{13}M_{31}}, \quad (3.28)$$

$$r_{34} = \left( \frac{E_{04}^{(0)}(z_0)}{E_{03}^{(0)}(z_0)} \right)_{E_{01}^{(0)}(z_0)=0} = \frac{M_{11}M_{43} - M_{41}M_{13}}{M_{11}M_{33} - M_{13}M_{31}}, \quad (3.29)$$

$$r_{32} = \left( \frac{E_{02}^{(0)}(z_0)}{E_{03}^{(0)}(z_0)} \right)_{E_{01}^{(0)}(z_0)=0} = \frac{M_{11}M_{23} - M_{21}M_{13}}{M_{11}M_{33} - M_{13}M_{31}}. \quad (3.30)$$

Similarly, we can express transmission coefficients as [9, 13]

$$t_{11} = \left( \frac{E_{01}^{(m+1)}(z_0)}{E_{01}^{(0)}(z_0)} \right)_{E_{03}^{(0)}(z_0)=0} = \frac{M_{33}}{M_{11}M_{33} - M_{13}M_{31}}, \quad (3.31)$$

$$t_{14} = \left( \frac{E_{03}^{(m+1)}(z_m)}{E_{01}^{(0)}(z_0)} \right)_{E_{03}^{(0)}(z_0)=0} = \frac{-M_{31}}{M_{11}M_{33} - M_{13}M_{31}}, \quad (3.32)$$

$$t_{33} = \left( \frac{E_{04}^{(m+1)}(z_m)}{E_{03}^{(0)}(z_0)} \right)_{E_{01}^{(0)}(z_0)=0} = \frac{M_{11}}{M_{11}M_{33} - M_{13}M_{31}}, \quad (3.33)$$

$$t_{31} = \left( \frac{E_{01}^{(m+1)}(z_m)}{E_{03}^{(0)}(z_0)} \right)_{E_{01}^{(0)}(z_0)=0} = \frac{-M_{13}}{M_{11}M_{33} - M_{13}M_{31}}. \quad (3.34)$$

For linearly polarized modes, it is possible to define these coefficients as the elements of Jones reflection matrices  $S_R$  and  $S_T$

$$S_R = \begin{bmatrix} r_{ss} & r_{sp} \\ r_{ps} & r_{pp} \end{bmatrix} = \begin{bmatrix} r_{12} & r_{32} \\ -r_{14} & -r_{34} \end{bmatrix}, \quad (3.35)$$

$$S_T = \begin{bmatrix} t_{ss} & t_{sp} \\ t_{ps} & t_{pp} \end{bmatrix} = \begin{bmatrix} t_{11} & t_{31} \\ t_{13} & t_{33} \end{bmatrix}. \quad (3.36)$$

and thus, determine the MO parameters  $\theta_K$ ,  $\varepsilon_K$  and  $\theta_F$ ,  $\varepsilon_F$ .

### 3.3. Proper modes in isotropic media

Permittivity of isotropic, in our case a non-magnetic environment, is a scalar and it is given by equation  $\varepsilon_I = N^{(n)2}$ . In this case, proper modes calculation procedure above leads to a solution with  $e_j$  components equal zero. Therefore, we calculate here with constant  $N_y$ . This approach determines two possible directions of the wave vector

$$\gamma_{1,2} = \frac{\omega}{c} \left[ N_y i_y \pm Q^{(n)} i_z \right], \quad (3.37)$$

where a parameter  $Q$  is calculated from

$$Q^{(n)} = \sqrt{N^{(n)^2} - N_y^2}. \quad (3.38)$$

This approach allows constructing the Dynamical matrix for isotropic environment in form [9]

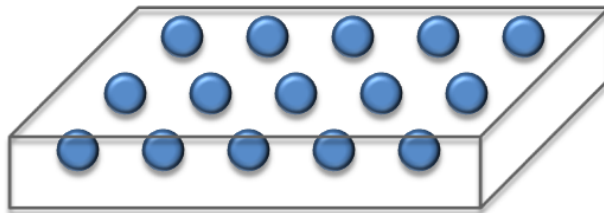
$$D^{(n)} = \begin{bmatrix} 1 & 1 & 0 & 0 \\ Q^{(n)} & -Q^{(n)} & 0 & 0 \\ 0 & 0 & Q^{(n)} / N^{(n)} & Q^{(n)} / N^{(n)} \\ 0 & 0 & -N^{(n)} & N^{(n)} \end{bmatrix} \quad (3.39)$$

and its inverse

$$[D^{(n)}]^{-1} = \begin{bmatrix} Q^{(n)} & 1 & 0 & 0 \\ Q^{(n)} & -1 & 0 & 0 \\ 0 & 0 & N^{(n)} & -Q^{(n)} / N^{(n)} \\ 0 & 0 & N^{(n)} & Q^{(n)} / N^{(n)} \end{bmatrix}. \quad (3.40)$$

### 3.4. Effective medium approximation

Interfaces and surfaces of real nanolayers are not perfect. Imperfections often contribute to the deviation between optical and MO experimental data versus theoretical models. The effective medium approximation method (EMA) can eliminate and quantitatively estimate this effect. The principle of this method lies in the consideration of the roughness or interface as a separate layer. This layer is constructed from two materials: the material of the first layer that forms interface and the material of the second layer that forms interface. Figure 3.2 shows geometric model of this approach. In this model is the material of the first layer dissolved in the form of spherical objects in the material of the second layer.



**Figure 3.2: Geometric model of the effective medium approximation method**



Here, we define permittivity tensor of the first layer that forms interface  $\varepsilon_I$  by its diagonal elements  $\varepsilon_{11}$  and off-diagonal elements  $\varepsilon_{12}$  and we rewrite it to the form  $\varepsilon_I = \varepsilon_{11} - i\varepsilon_{12}$ . Similarly, we can define the permittivity tensor of the second layer that forms interface  $\varepsilon_2 = \varepsilon_{21} - i\varepsilon_{22}$  by diagonal elements  $\varepsilon_{21}$  and off-diagonal elements  $\varepsilon_{22}$ . Now, we can use our geometric model (Figure 3.2) and define the interface layer permittivity tensor by diagonals  $\varepsilon_{R1}$  and off-diagonals  $\varepsilon_{R2}$  using transformation relations

$$\varepsilon_{R1} = \varepsilon_{12} + f \frac{\varepsilon_{11} - \varepsilon_{12}}{1 + \frac{(1-f)(\varepsilon_{11} - \varepsilon_{12})}{3\varepsilon_{12}}} \quad (3.41)$$

$$\varepsilon_{R2} = f \frac{\varepsilon_{21}}{1 + \frac{(1-f)(\varepsilon_{11} - \varepsilon_{12})}{3\varepsilon_{12}}}. \quad (3.42)$$

Coefficient  $f$  corresponds to the volume fraction of the second material forming the interface (for example,  $f = 0.5$  corresponds to 50% of the second material in the mixture) [15].

For the case of the surface roughness calculations, we assume the second material to be void. If we rewrite the permittivity tensor of the layer material to the form  $\varepsilon_I = \varepsilon_{11} - i\varepsilon_{12}$  then we can calculate permittivity tensor of the surface roughness layer  $\varepsilon_R = \varepsilon_{R1} - i\varepsilon_{R2}$  using equation

$$0 = (1-f) \frac{\varepsilon - \varepsilon_R}{\varepsilon + 2\varepsilon_R} + \frac{1 - \varepsilon_R}{1 + 2\varepsilon_R} \quad (3.43)$$

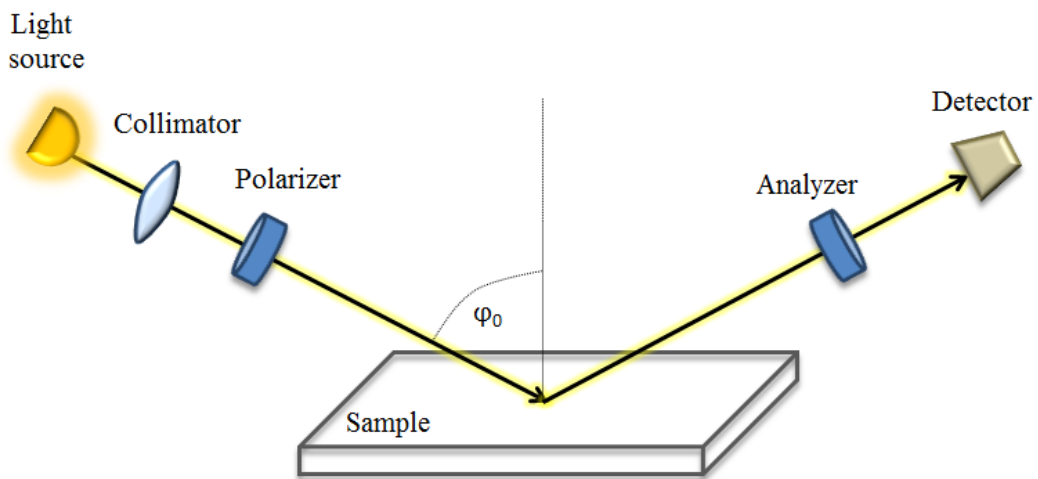
In this formula,  $f$  denotes the volume fraction of the void in the mixture.

## 4. EXPERIMENTAL TECHNIQUES

In this chapter we discuss briefly experimental techniques used for acquisition of experimental data in this work. We start this chapter with a brief introduction to spectroscopic ellipsometer, which is used for measurements of thin film optical properties and allows determination of thin film optical functions (complex permittivity tensor). Afterwards, we continue with an introduction to measurements of weak MO effects: MOKE and Faraday effect. Spectra of MO effects contain important information about electronic structure of thin films and allow determination of the full permittivity tensor.

### 4.1. Spectroscopic ellipsometer

As explained previously, SE measures changes in the polarization state of light upon reflection or transmission on the sample. In this sub-chapter, we discuss basic setup for this type of measurement.



**Figure 4.1: Scheme of the basic ellipsometry setup**

All spectroscopic ellipsometer arrangements start with a light source and end with a detector that converts light into voltage. It is an arrangement of the optical components between that defines the type of ellipsometer being used. In this work, we use Muller matrix ellipsometer Woollam RC2 with dual rotating compensators

and multichannel detection working in the photon energy range from 1.24 to 6.5 eV. Further details on this complicated device can be found in [2, 12]. However, for our purposes, here we discuss only the basic set-up of ellipsometer shown in the Figure 4.1. It is constructed from a rotating polarizer that rotates on frequency  $\omega$ , sample and analyzer. For this type of setup and general elliptical polarization, one can determine alternating current signal on detector  $I$  as

$$I \approx 1 + \alpha \cdot \cos(2\omega t) + \beta \cdot \sin(2\omega t) \quad (4.1)$$

The two important quantities are  $\alpha$  and  $\beta$ , which are normalized Fourier coefficients of the signal. One can represent these coefficients in terms of the  $\psi$  and  $\Delta$  defined in the sub-chapter 2.2 and the known polarizer azimuthal angle  $P$  as follows ( $P=0^\circ$  is in the plane of incidence)

$$\alpha = \frac{\tan^2 \psi - \tan^2 P}{\tan^2 \psi + \tan^2 P} \quad (4.2)$$

$$\beta = \frac{2 \tan \psi \cos \Delta \tan P}{\tan^2 \psi + \tan^2 P} \quad (4.3)$$

Solution for  $\psi$  and  $\Delta$  has therefore a form

$$\tan \psi = \sqrt{\frac{1+\alpha}{1-\alpha}} |\tan P|, \quad (4.4)$$

$$\cos \Delta = \frac{\beta}{\sqrt{1-\alpha^2}} \cdot \frac{\tan P}{|\tan P|}. \quad (4.5)$$

These equations form the basis of the ellipsometry measurement with a rotating analyzer ellipsometer [12].

For experimental data analyses, we use a CompleteEASE software. This software compares the data acquired by the ellipsometer with an advanced theoretical model of studied multilayer. Here, theoretical model describes the multilayer as a structure that consists of separate layers. These separate layers are determined by their optical functions (complex permittivity), thicknesses and by their positions in the multilayered structure.

In this work, we use “**Multi Sample Analysis**” (MSA) mode in the CompleteEASE software multiple times. MSA is an advanced mode that allows simultaneous fit of data from multiple samples. MSA enable to set some of fit parameters common to all samples and other parameters are allowed to vary (optical functions of materials, thicknesses, roughness, backside reflections, angle offset,...) [2]. Therefore, this CompleteEASE feature suppresses the fit error and allows more precise analysis. The MSA mode is especially useful, when combining the reflection and transmission data. In our case, we use MSA to fit one theoretical model to the ellipsometry and transmission data simultaneously. This approach is especially meaningful when fitting constants of transparent materials with a bandgap in the measured spectral range.

## 4.2. Magneto-optical spectroscopy

The simplest way to define MO spectroscopy is to say that it is SE performed in the magnetic field. Therefore, MOKE spectroscopy is basically SE which measures a change in the polarization state of light upon reflection on the sample in magnetic field. Similarly, Faraday effect spectroscopy is SE which measures a change in the polarization state of light upon transmission on the sample in magnetic field. These types of measurements are useful for non-destructive probing of magnetic properties of non-transparent (MOKE) or transparent (Faraday) magnetic materials and nanostructures. Since some materials have absorption edge within measured spectral range, it is a classical approach to combine these measurements for different parts of spectra.

In this work, we measure MOKE by method of nearly crossed polarizers. Figure 4.2 shows basic scheme of this method: the light beam is passing through the polarizer, reflecting on the sample in magnetic field, passing through the phase plate and finally through the analyzer to the detector. For this type of MOKE setup and general elliptical polarization, one can determine intensity on the detector  $I$  as

$$I \approx \cos^2 \gamma + |\Phi_K|^2 \sin^2 \gamma + \sin(2\gamma) \Re(\Phi_K e^{i\delta}) \quad (4.6)$$

Here,  $\gamma$  is an angle between analyzer and polarizer with respect to the crossed position and  $\delta$  is a phase shift of the phase plate.

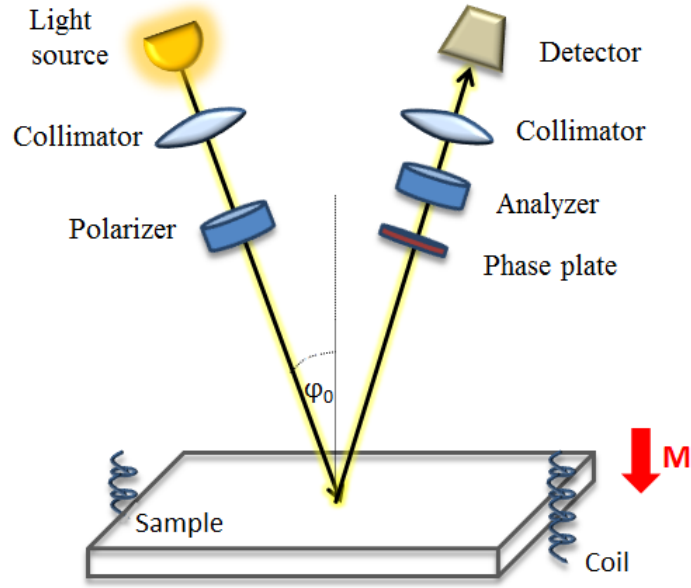


Figure 4.2: Scheme of the basic MOKE setup

If we restrict ourselves to effects linear in  $\Phi_K$ , and add constant  $C$  corresponding to a dark current in CCD, we can rewrite resulting dependence of detected intensity on the angle of analyzer  $\alpha$  as

$$I \approx \cos^2 \gamma + (\theta_K \cos \delta + \epsilon_K \sin \delta) \sin 2\gamma + C \quad (4.7)$$

Since it is complicated to measure Kerr rotation  $\theta_K$  and Kerr ellipticity  $\epsilon_K$  in one measurement, it is usually measured separately. This is done by removing a phase plate, which results in  $\delta=0$ , therefore, we measure pure Kerr rotation. To extract ellipticity, we combine experiments with and without phase plate. For more details, one can see [16].

The basic setup for Faraday effect measurement can be seen in Figure 4.3: the light beam is passing through the polarizer, through the sample in magnetic field and finally through the analyzer to the detector. For this type of setup, calculations lead to the same expression that it is for MOKE, except from the change in sign of term with MO effect.

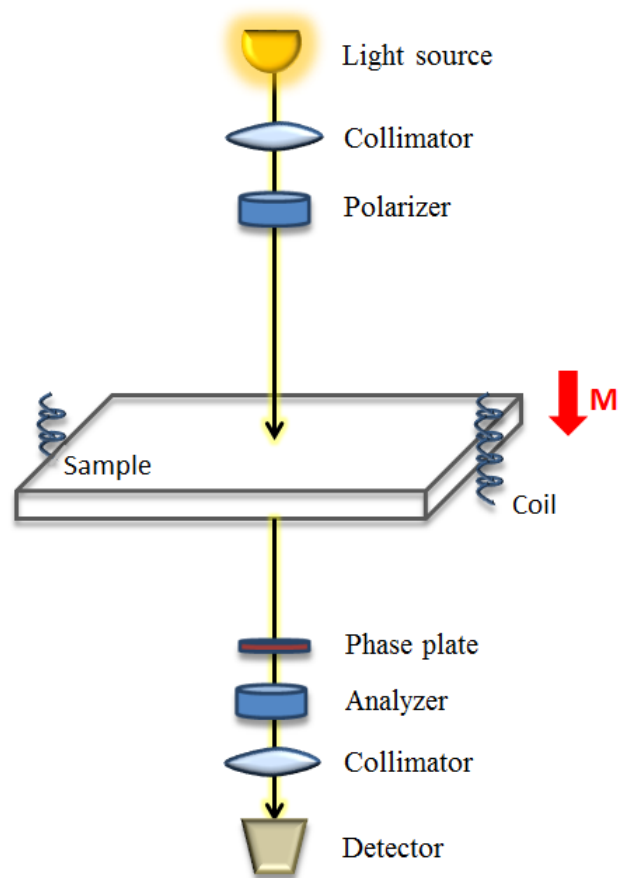


Figure 4.3: Scheme of the basic Faraday effect setup.

## 5. THIN FILM PREPARATION TECHNIQUES

In the previous chapter we discuss methods used for acquirement of experimental data. Correspondingly, we devote this chapter to techniques employed for the preparation of materials/samples studied in this work. These preparation techniques are relatively new, require advance technology and sometimes even allow preparation of bulk materials, thin films and nanostructures which do not normally exist in nature.

### 5.1. Sputter deposition

Sputter deposition is a thin films preparation method that involves bombardment of the material source (target) by energetic particles. This results in the sputtering of its atoms into the gas phase. Since these atoms are not in their thermodynamic equilibrium state, they tend to deposit on all surfaces in the vacuum chamber. Therefore, a substrate placed in the chamber is coated with a thin film.

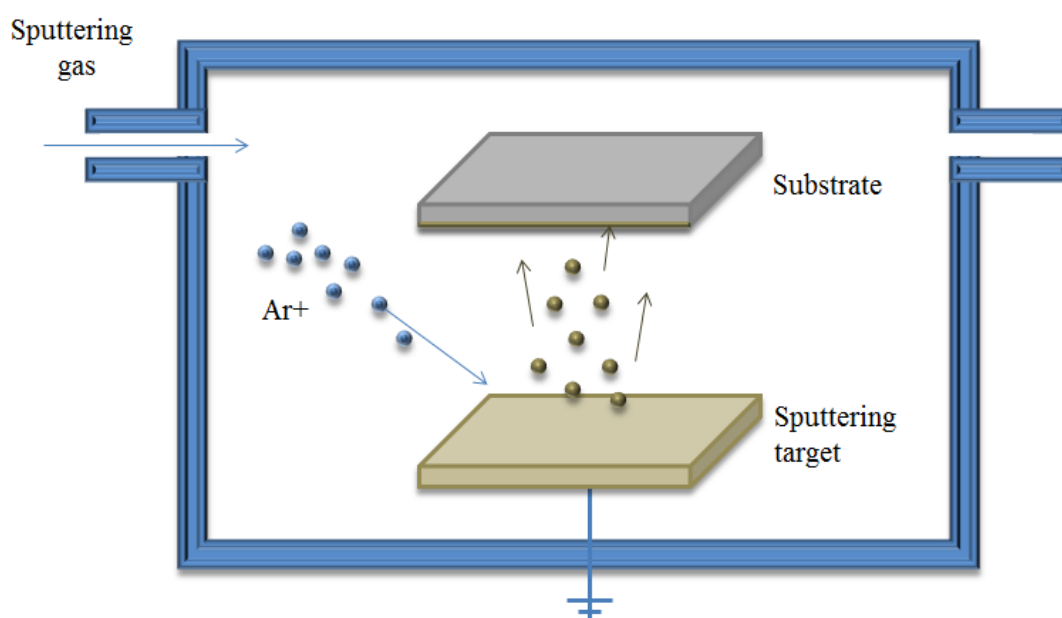


Figure 5.1: Sputter deposition.

**Direct Current or DC Sputtering** [17] is the most basic and inexpensive technique for deposition of metals or electrically conductive coating materials. Major

advantages of this technique are easy control due to DC source and low cost (when preparing metallic films). The basic configuration of a DC Sputtering coating system is schematically shown in the Figure 5.1. It consists of the target material placed in a vacuum chamber parallel to the substrate. The vacuum chamber is evacuated to a low pressure and subsequently backfilled with a high purity inert gas. This inert gas is usually Argon with typical sputter pressure range from 0.5mTorr to 100mTorr. Argon is used due to its relative mass and ability to convey kinetic energy within high energy molecular collisions in the plasma. Afterwards a DC current is applied to the target which serves as a cathode or negative bias (point where electrons enter the system). Subsequently, a positive charge is applied to the substrate which serves as an anode or positive bias. Firstly, the electrically neutral argon gas atoms are ionized due to the collisions with the negatively charged target. This ejects atoms off into the plasma. Secondly, the ionized argon atoms are driven to the anode substrate, which is attracting ionized gas ions, electrons, and the most importantly vaporized target material atoms. Finally, these atoms condense into a thin film coating on the substrate. While DC Sputtering is a great choice for preparation of multiple conductive coatings, its main limitation lies in non-conducting dielectric insulating materials. These take on a charge over time which usually results in quality issues like arcing, poisoning of the target material with a charge and even in the complete cessation of sputtering. To overcome these limitations, several technologies such as radio frequency sputtering have been developed.

**Radio Frequency or RF Sputtering** [17] is very similar to the DC sputtering technique. The difference lies in the alternating of the current electrical potential in the vacuum environment at radio frequencies. This is done to avoid a charge build up on certain types of target material. The charge buildup is cleaned every positive cycle when electrons are attracted to the target giving it a negative bias. During the negative cycle, ion bombardment of the target continues normally. The alternating of the current electrical potential is performed by a capacitor. The capacitor is a part of impedance-matching network. This network conveys the power transfer from the RF source to the plasma discharge. While RF Sputtering is extremely useful technique, it also has several disadvantages. For example, when using radio waves instead of DC current, deposition rates became considerably slower. Moreover, this technique requires significantly higher voltages (expensive power supplies requirement) to

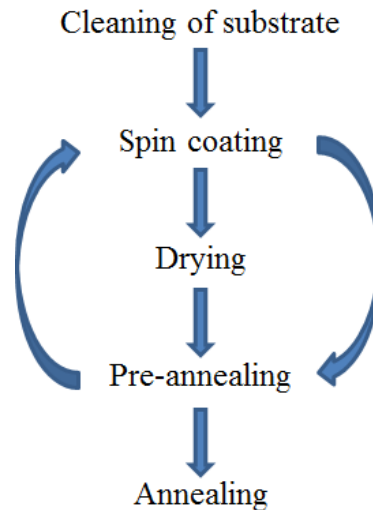


achieve the same deposition results as with DC, and so overheating (advanced circuitry requirement) also becomes an issue.

## 5.2. Metal Organic Decomposition

Metal Organic Decomposition (MOD) is a technique for manufacturing of inorganic thin films. The major advantage of this technique is that manufacturing process does not involve processing in the vacuum or going through a gel or powder step [18]. The key to this method are metal-organic components. These are prepared by dissolving required element in an appropriate solvent. Metal-organic components are mixed in an appropriate ratio to give a metal-organic solution which results in desired cation stoichiometry for the final film. The simplified process of MOD can be seen in Figure 5.2. The first step is a deposition and spin coating of a metal-organic solution on a substrate and therefore a production of a wet film. Afterwards, the film is heated multiple times (drying, pre-annealing). This is done to remove any solvent that did not evaporate during the deposition step and to decompose the metal-organic compounds. At the end, this process results into inorganic film of some thickness. This thickness is constant when all the steps are performed under the same conditions. If the inorganic film produced by a single pass through the process is not thick enough, all the steps can be repeated as many times as necessary to produce a film of the required thickness. As soon as the desired film thickness is achieved, the film is heated one more time (annealing) to control features such as oxygen stoichiometry, grain size or preferred orientation.

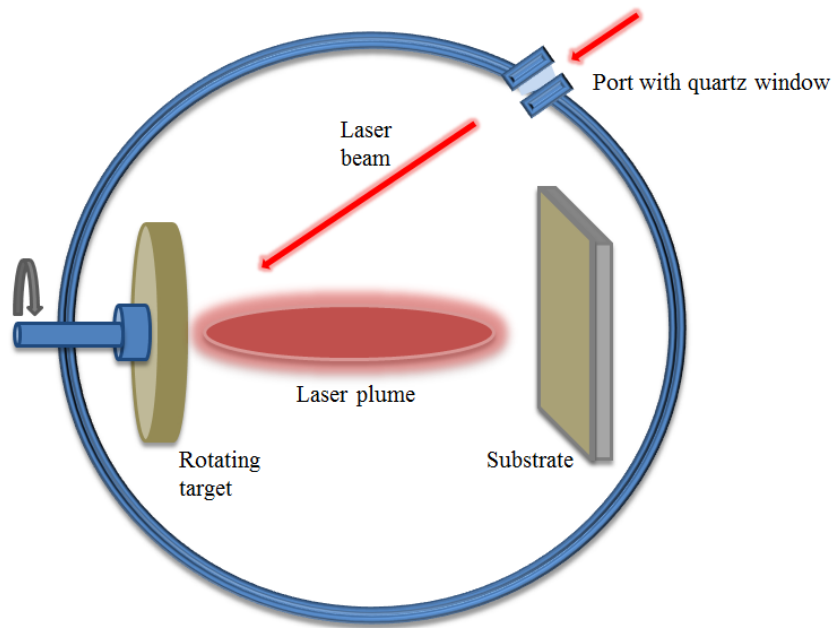
Metal organic decomposition is a very promising method since it has multiple advantages. First of all, it is inexpensive, guarantees highly uniform chemical composition and purity combined with a good chemical stability [19, 20]. Furthermore, it provides a good productivity, since it involves simple processes performed in the air. MOD also allows epitaxial growth of thin (mostly garnet) films on lattice constant mismatching substrates, which is not possible by techniques such as liquid phase. Finally, MOD ensures a possibility of a thin film formation over a large area.



**Figure 5.2: Simplified process of MOD**

### **5.3. Pulsed laser deposition**

In principle, pulsed laser deposition [21, 22] is a simple technique schematically shown in Figure 5.3. This technique uses high-power pulsed laser beam (typically  $\sim 108 \text{ Wcm}^{-2}$ ) and focuses it into a vacuum chamber to strike a target of a deposited material. This striking event produces plasma plume that augments rapidly away from the target surface. The struck material is then collected on a substrate where it condenses and grows as a thin film. This process usually occurs in ultra-high vacuum. However, sometimes a background gas, such as oxygen is used when depositing oxides to fully oxygenate the deposited films. In practice, there are multiple variables affecting properties of the film (laser fluence, background gas pressure, substrate temperature etcetera). Therefore, the process optimization requires a lot of time and effort. Application span of this technique is wide. It ranges from the production of superconducting and insulating circuit components to various medical applications. Unfortunately, the fundamental processes occurring during the transfer of material from target to substrate are still not clear and are consequently the focus of much research.



**Figure 5.3: Pulsed laser deposition**

## 6. RESULTS

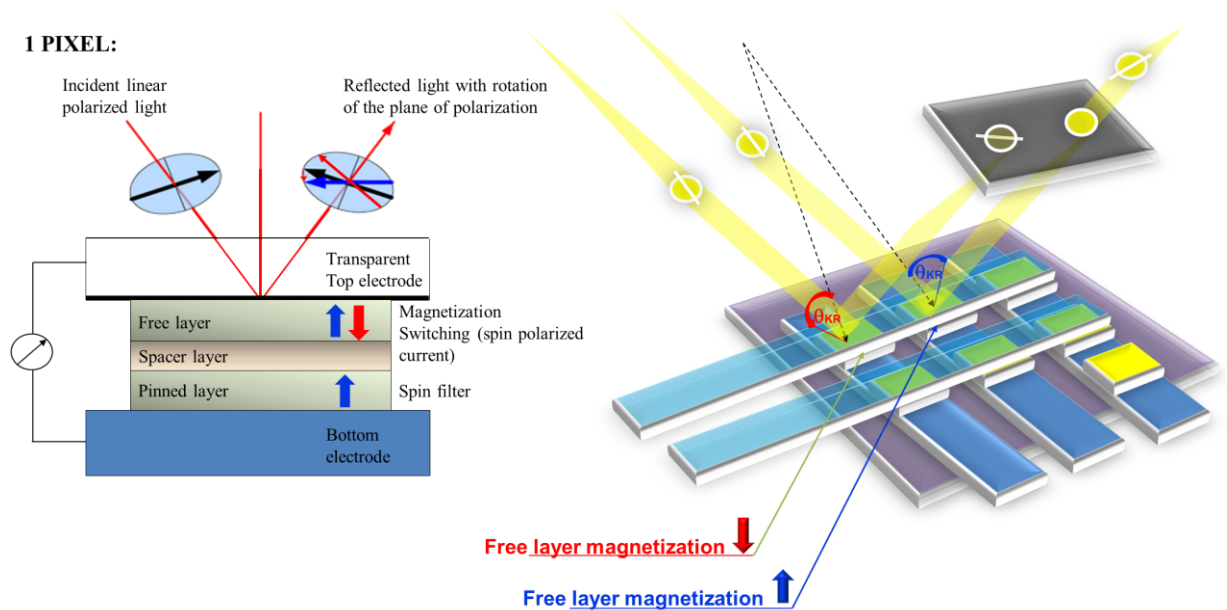
In this chapter, we discuss in detail multiple materials with application potential in several optical and MO devices. We structure this chapter in sub-chapters which are devoted to individual materials:  $\text{Gd}_x\text{Fe}_{(100-x)}$ , magnetic garnets and  $\text{Ce}_{(0.95-x)}\text{Hf}_x\text{Co}_{0.05}\text{O}_{(2-\delta)}$ . At the beginning of every sub-chapter, we discuss the application potential of the material, its properties, samples preparation and samples structure. Afterwards, we focus on SE measurements and analysis. Finally, for magnetic materials, we focus on MO spectroscopy (MOKE and Faraday effect) measurements and analysis.

### 6.1. $\text{Gd}_x\text{Fe}_{(100-x)}$

Amorphous ferrimagnetic thin films composed of rare earth and transition metals attracted considerable attention because of their useful technological applications [23-26]. As one of their important representative,  $\text{Gd}_x\text{Fe}_{(100-x)}$  has significant advantages, such as large magnetization density, and possibility to adjust its compensation temperature, coercive and saturation magnetization by changing the composition [27-29]. Another valuable feature of  $\text{Gd}_x\text{Fe}_{(100-x)}$  is that it enables direct access to its spins through the electromagnetic interactions. Such feature makes this material subject of importance for future magnetic recording (such as heat assisted magnetic recording) and information processing technologies. Recent numerical atomic scale modeling simulations of the spin dynamic in Heisenberg  $\text{Gd}_x\text{Fe}_{(100-x)}$  ferrimagnet demonstrated that the rapid transfer of energy into the spin system leads to switching of the magnetization within a few ps without necessity of applied magnetic field. The experiment in  $\text{GdFeCo}$  alloys, which used linearly polarized fs laser pulse to produce the ultra-fast heating, confirmed this prediction [30-32]. Moreover, by using circularly polarized laser pulses, it is possible to take an advantage of the magnetic circular dichroic effect to record a magnetic domain in which the helicity of the laser pulse influences the final magnetization direction [30, 32-34]. These mechanisms allow the  $\text{Gd}_x\text{Fe}_{(100-x)}$  magnetic domain light spin manipulation and hence coherent control of the magnetization precession at fluencies as low as  $6 \mu\text{J}/\text{cm}^2$  [35] and in rates of ps [32, 33, 36]. All of mentioned properties make  $\text{Gd}_x\text{Fe}_{(100-x)}$  substantial material for modern micro- and nano-electronic

research, where it is often used in domain wall junctions or magneto-optical (MO) memories [23, 25, 26].

Recently, a novel concept of high speed MO spatial light modulator (MO-SLM) for holographic displays based on giant magnetoresistance with  $Gd_xFe_{(100-x)}$  as a free layer was proposed [24]. This device is shown in Figure 6.1.1. As one can see, every pixel consists of the ‘free magnetization layer (FL)/ nonmagnetic spacer/ pinned magnetization layer (PL)’ structure. Free layer magnetization is controlled by spin polarized current. When the magnetizations of FL and PL are aligned, the polarization of the light reflected on the structure will rotate to the one direction. When magnetizations of FL and PL head against each other, the polarization of the light reflected on the structure will rotate to the opposite direction. Therefore, in the multi-pixel structure, we can create interference pattern crucial for holography, just by using a polarization filter. The main advantage of this approach is the response time in terms of  $0.015 \mu s$  and pixel size in terms of  $10 \mu m$  [37].



**Figure 6.1.1: Concept of high-speed MO spatial light modulator for holographic displays based on GMR.**

When using  $Gd_xFe_{(100-x)}$  for MO-SLM driven by spin transfer torque, it is particularly important that  $Gd_xFe_{(100-x)}$  shows perpendicular anisotropy. This happens when the Fe concentration is close to the compensation concentration, which is for this

material about 75% [38] (this composition is also often used for MO applications such as MO disk storage [24]). Since in here  $Gd_xFe_{(100-x)}$  acts as FL, it is especially important to control its composition precisely, since it significantly affects its magnetic switching property. Coercivity shows maxima when the  $Gd_xFe_{(100-x)}$  composition is the compensation one, and it gets smaller when the composition becomes Fe rich (compared to the compensation composition). Spin-torque switching current of the spin MO-SLM is significantly reduced with an increase in Fe concentration and it shows very small switching current when composition is slightly Fe richer than the compensation one [37, 39]. Therefore, it is meaningful to investigate optical properties of the  $Gd_xFe_{(100-x)}$  material around the compensation concentration.

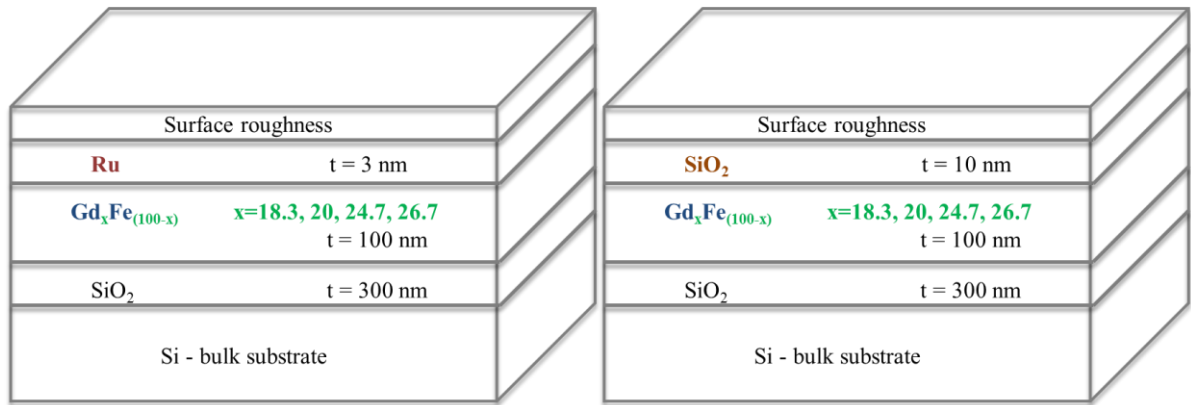
The main purpose of our investigation was the determination of complete permittivity tensors for  $Gd_xFe_{(100-x)}$  thin films with various compositions ( $x=18.3, 20, 24.7, 26.7$ ). We were interested in the permittivity tensor spectra because it provides a deeper look into optical and MO properties of material. Moreover, the knowledge of the complete tensor allows the theoretical prediction of complex physical properties of complicated multilayered nanostructures containing  $Gd_xFe_{(100-x)}$  layer. This is especially useful when designing complicated nanostructured device such as MO-SLM, because it allows proposal of desired structure without a necessity of preparing and measuring multiple samples.

In this work is  $Gd_xFe_{(100-x)}$  covered by a coating to avoid the oxidation process [40]. However, this fact complicates its analysis. Optical properties of coating materials (here Ru,  $SiO_2$ ) may slightly differ in dependence on material they are deposited on. The reason behind this behavior is usually the lattice mismatch between the film and substrate. This mismatch induces strains of various kinds [41-43]. To deal with this problem, we used 2 different coating materials. Such approach allowed more precise determination of  $Gd_xFe_{(100-x)}$  permittivity tensors. SE analysis showed remarkably similar optical properties of individual  $Gd_xFe_{(100-x)}$  compositions for both coatings, which demonstrated a good stability of  $Gd_xFe_{(100-x)}$  layer.

For the measurement of  $Gd_xFe_{(100-x)}$  properties, we used SE at energies from 1.3 to 5.5 eV and MO spectral measurements at energies from 1.5 to 5.5 eV. From SE data, we derived the diagonal permittivity tensor elements  $\epsilon_{1r}$  and  $\epsilon_{1i}$  spectra of  $Gd_xFe_{(100-x)}$

thin films. We examined MO properties by polar MOKE rotation and ellipticity measurements. From these data we determined the spectral dependence of the off-diagonal  $Gd_xFe_{(100-x)}$  permittivity tensor elements  $\epsilon_{2r}$  and  $\epsilon_{2i}$ . We also performed MOKE hysteresis measurements, which demonstrated changes in the magnetization in dependence on  $Gd_xFe_{(100-x)}$  composition.

For our samples, we used silicon substrate with thermally oxidized  $SiO_2$  layer. Afterwards, we deposited  $Gd_xFe_{(100-x)}$  and Ru coating by direct current sputtering technique in Kr gas of pressure  $8.7 \times 10^{-2}$  Pa with a deposition rate of 3.6 nm/min. Finally, we deposited  $SiO_2$  coating by ion beam sputtering technique with radio frequency ion source. Theoretical model structures used for SE and MOKE analysis calculations are shown in Figure 6.1.2.



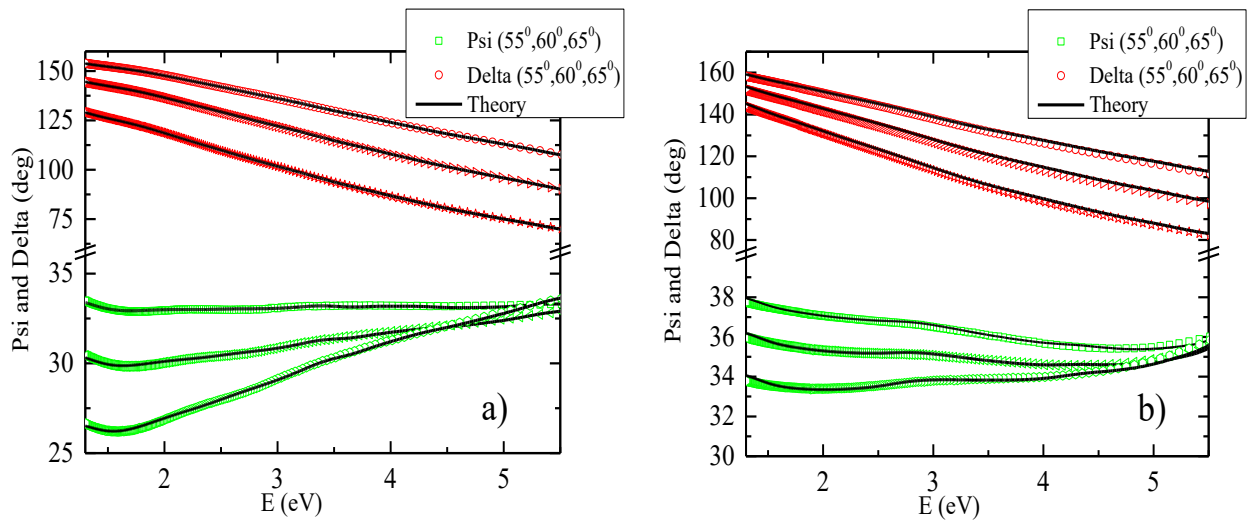
**Figure 6.1.2: Model structure of GdFe/Ru and GdFe/ $SiO_2$  samples used for SE and MOKE analysis calculations.**

### a) Spectroscopic Ellipsometry

We measured ellipsometry Psi and Delta parameters of the reflected light in the spectral range from 1.3 to 5.5 eV for incident angles  $55^\circ$ ,  $60^\circ$  and  $65^\circ$ . Obtained experimental data were analyzed using CompleteEase software. To analyze SE experimental data of studied samples we fitted  $Gd_xFe_{(100-x)}$  optical functions (diagonal elements of the permittivity tensor  $\epsilon_{1r}$  and  $\epsilon_{1i}$ ) and nominal thicknesses to the structural models and experimental data. In order to avoid the false minima result of the fitting process, we used the MSA mode to derive the  $Gd_{18.3}Fe_{81.7}$  and  $Gd_{24.7}Fe_{75.3}$  optical functions from the experimental data for both coatings

simultaneously. In MSA mode, we set  $Gd_xFe_{(100-x)}$  ( $x=18.3$  or  $x=24.7$ ) optical functions as the fit parameter common for both samples. We set optical functions of coating materials and thicknesses as fit parameters varying for each sample independently. Figure 6.1.3 shows that theoretical model describes both:  $Gd_{20}Fe_{80}/Ru$  and  $Gd_{20}Fe_{80}/SiO_2$  SE experimental spectra well.

We parameterized obtained optical functions to ensure Kramers-Kronig consistent results. We used two Lorentz oscillators and one Drude term in the spectral range from 1.5 to 6 eV. We used Lorentz oscillators due to their ability rapidly approach zero beyond the FWHM position. We used Drude term to describe the free carrier effect on the dielectric response. We listed parameters of used parameterizations in the table 6.1.1. We determined constants of Si,  $SiO_2$  and Ru from SE measurements on individual samples. We listed derived thicknesses in the table 6.1.2. To confirm thicknesses of  $Gd_xFe_{(100-x)}$  layers derived from SE, we also performed XRF measurements. We listed thicknesses of  $Gd_xFe_{(100-x)}$  layers derived by XRF in the table 6.1.2. As you can see, values derived from SE and XRF correspond well.



**Figure 6.1.3: Measured variable angle SE Psi and Delta spectra of a)  $Gd_{20}Fe/Ru$  and b)  $Gd_{20}Fe/SiO_2$  samples compared to the theoretical model .**



**Table 6.1.1: Fitted parameters of Lorentz oscillators and Drude term used to parameterize optical functions of  $Gd_xFe_{(100-x)}$  in the spectral range from 1.5 to 6 eV. In here, E stands for central energies of oscillators; Amp represents amplitudes and Br broadenings. For Drude model, N represents carrier concentration,  $\mu$  carrier mobility and  $m^*$  carrier effective mass.**

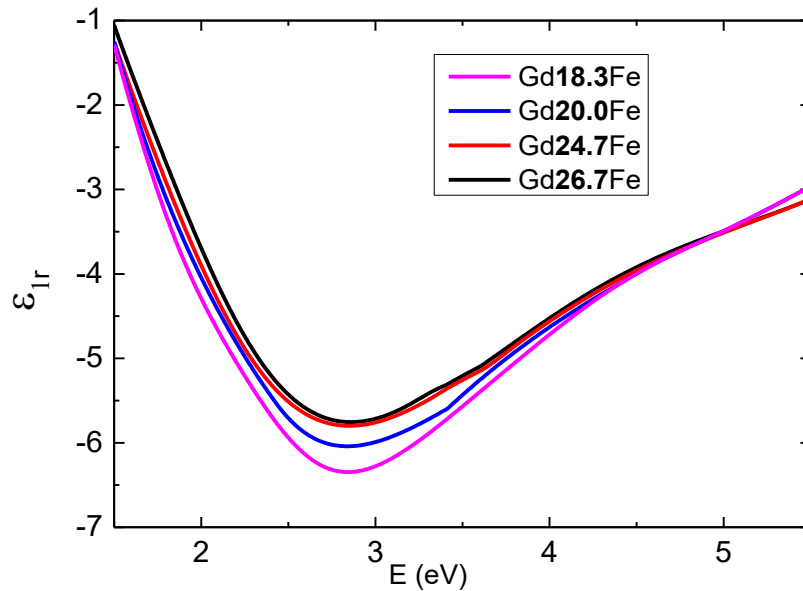
	Lorentz (E = 1.89 eV)		Lorentz (E = 2.57 eV)		Drude term		
	Amp	Br (eV)	Amp	Br (eV)	N (cm <sup>-3</sup> )	M (cm <sup>2</sup> V <sup>-1</sup> s <sup>-1</sup> )	$m^*$
Gd18.3Fe	6.66	2.30	1.10	1.09	1.098*10 <sup>23</sup>	0.354	0.514
Gd20.0Fe	6.23	2.43	1.28	1.43	1.095*10 <sup>23</sup>	0.352	0.533
Gd24.7Fe	6.04	2.69	1.43	1.84	1.110*10 <sup>23</sup>	0.340	0.564
Gd26.7Fe	5.82	2.83	1.67	1.99	1.113*10 <sup>23</sup>	0.332	0.573

**Table 6.1.2: Fitted thicknesses used for model of  $Gd_xFe_{(100-x)}$  in SE and XRF in the spectral range from 1.5 to 6 eV. In here, t stands for thickness and r for roughness on top.**

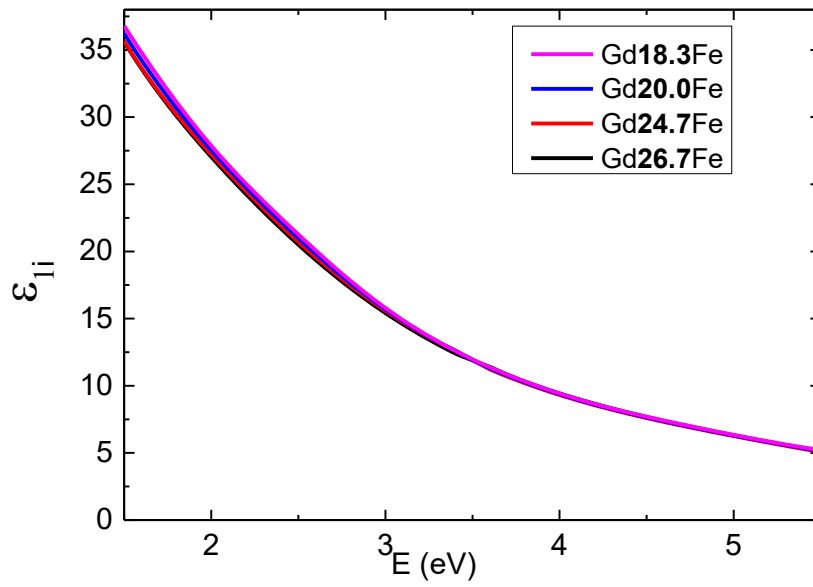
	$t_{SiO_2}$ (nm) SE	$t_{GdFe}$ (nm) SE	$t_{GdFe}$ (nm) XRF	$t_{Ru}$ (nm) coating	$t_{SiO_2}$ (nm) coating	r (nm) SE
Gd18.3Fe/Ru	307	131.6	136.9	3.1	--	2
Gd18.3Fe/SiO <sub>2</sub>	307	130	136.9	--	11.3	0.3
Gd20.0Fe/Ru	307	103	99.6	2.9	--	2
Gd24.7Fe/Ru	307	95	87.7	2.7	--	1.9
Gd24.7Fe/SiO <sub>2</sub>	307	87.7	87.7	--	10.5	0.7
Gd26.7Fe /Ru	307	93.4	93.4	2.2	--	1.9

Figure 6.1.4 shows real parts of the diagonal permittivity tensor elements  $\epsilon_{1r}$  of the  $Gd_xFe_{(100-x)}$  thin films. Figure 6.1.5 shows imaginary parts of the diagonal permittivity tensor elements  $\epsilon_{1i}$  of the  $Gd_xFe_{(100-x)}$  thin films. Figure 6.1.6 shows their calculated absorption coefficient spectra. The  $\epsilon_{1r}$  spectra are characteristic by one global minimum at 2.9 eV while the  $\epsilon_{1i}$  amplitudes decrease their values with increasing energy in the measured spectral range for all the compositions. As expected, absorption coefficient spectra have increasing character with energy. All results show spectral behavior similar to previously published optical properties of Fe, Gd [44, 45] and  $Gd_xFe_{(100-x)}$  films [28, 46]. The behavior in the spectral range from 1.5 to 3eV, where  $\epsilon_{1r}$  decreases its value for higher energies, differs from

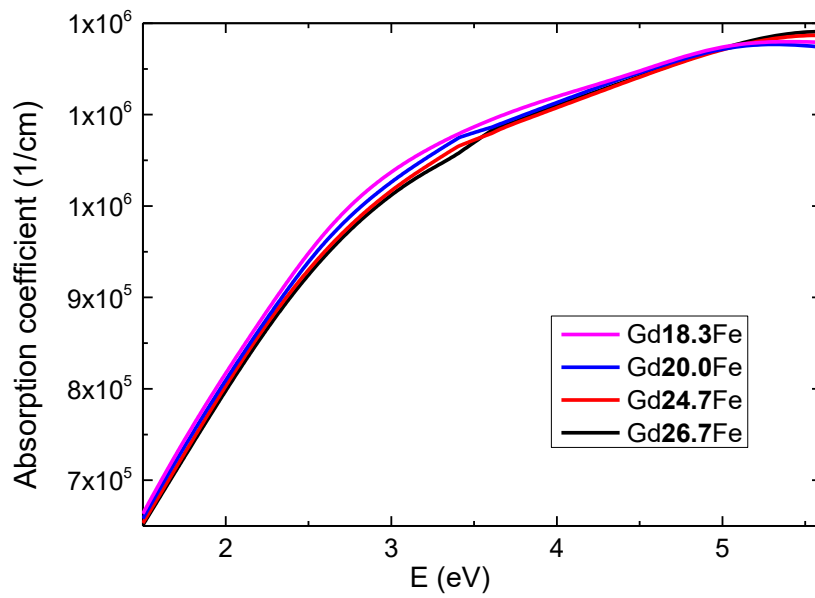
typical Drude-like behavior (describing intra-band transitions) of metallic compounds, and is similar to the behavior of some transition metals (including Cr, Gd, Ru, Ti [44, 45]). This behavior is caused by the Lorentz contribution centered near 1.9 eV. It most likely originates in the inter-band transition, which involves Fe 3d and Gd 5d states. The Fe 3d state is located around 1.5 eV below Fermi energy, while Gd 5d states are located approx. 0.5 eV above the Fermi energy [47, 48]. The second Lorentz oscillator near 2.5 eV does not significantly modify the spectral dependence of Drude behavior. The reason is its small amplitude. This points on the origin of Gd d-d electron transition [47, 48], since this transition should be forbidden with small oscillator strength. Finally, we discuss the Gd substitution effect. The amplitude of the first Lorentz oscillator near 1.9 eV decreases with Gd content. This is visible in Figure 6.1.4 as a change of  $\epsilon_{1r}$  amplitude around 2.9 eV. Increasing Gd content means decreasing Fe density of states below Fermi energy. This results in the suppression of Fe 3d to Gd 5d transition probability. On the contrary, the increase of Gd content is increasing the amplitude of the second Lorentz oscillator centered near 2.5 eV. Such behavior supports the assignment of the origin to Gd d-d transition.



**Figure 6.1.4: Real parts of diagonal elements of the permittivity tensor of the  $Gd_xFe_{(100-x)}$  thin films.**



**Figure 6.1.5: Imaginary parts of diagonal elements of the permittivity tensor of the  $Gd_xFe_{(100-x)}$  thin films.**



**Figure 6.1.6: Calculated absorption coefficients of  $Gd_xFe_{100-x}$  thin films.**

## b) MOKE spectroscopy

We measured MO properties by MOKE spectroscopy. We obtained the MOKE rotation and ellipticity spectra in the polar configuration. We acquired all the spectra at the room temperature for nearly normal light incidence. Applied magnetic field was 1.2 T, which was enough for magnetic saturation of samples. Incident light was *s* polarized. We recorded data in the photon energy range from 1.5 to 5.5 eV. We measured MOKE rotation hysteresis loops by differential intensity detection method at 2.38 eV. We performed all measurements in the polar geometry and at the room temperature. Field was ranging from -1.8 T up to 1.8 T, which was far beyond saturation point.

We would like to start this section with polar MOKE hysteresis measurements shown in Figure 6.1.7. First thing to notice is that samples with higher Gd content exhibit square-like loops with sharp transition. This indicates out-of-plane easy axis of  $\text{Gd}_x\text{Fe}_{(100-x)}$  net magnetization for all the samples. As the composition approaches the compensation point, the coercivity increases. Below the composition value of  $x \approx 25$ , the Fe moments become more dominant forcing the sample accommodate the in-plane anisotropy. The higher net magnetization value results in stronger effects of dipolar interaction, inducing a complex multi-domain state in the sample with negligible coercivity. With lower Gd content, the hysteresis loop shape becomes more prolonged and together with the increase of the saturation field hints on tilted direction of the net magnetization to the OOP orientation [49]. Such state is described by complex butterfly-like shaped hysteresis loops shown in Figure 6.1.7 for samples Gd18.3Fe and Gd20Fe. Further thing to discuss, is the opposite direction of the magnetization for samples with the Gd concentration  $x=24.7$  (Gd24.7Fe/Ru and Gd24.7Fe/SiO<sub>2</sub>). Since this concentration is extremely close to the compensation point, it would be reasonable to assume that the dispersion of the Gd concentration during the film preparation process causes this behavior. However, we believe that in this case (based on experience), coating affects the Gd24.7Fe properties more than Gd concentration dispersion, since SiO<sub>2</sub> may oxidize Gd selectively.

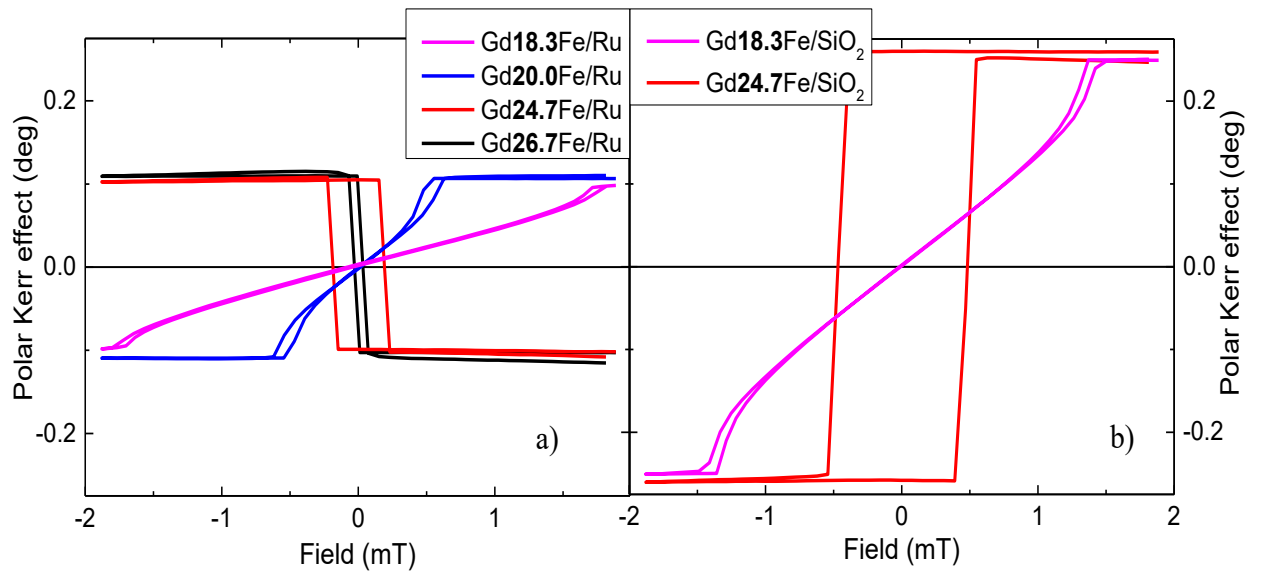


Figure 6.1.7: Hysteresis loops of examined samples with a) Ru and b) SiO<sub>2</sub> coatings at 2.38 eV.

Figure 6.1.8 shows MOKE rotation and Figure 6.1.9 MOKE ellipticity spectra. Firstly, both spectra are characteristic by increasing rotation and ellipticity amplitudes towards to smaller energies. Secondly, samples with SiO<sub>2</sub> coating show much higher MO signal than samples with Ru coating (also possible to see from hysteresis loops measurements). We attributed this to multiple reflections in measured energy region for SiO<sub>2</sub> coated samples. Furthermore, we can observe that substitution of Gd is increasing amplitudes of MOKE. Moreover, as expected, amplitudes of MOKE rotation and ellipticity changes the sign when Gd reaches the compensation concentration ( $x \approx 25$ ) and therefore when net magnetization direction changes. All the data correspond with the hysteresis loops measurements.

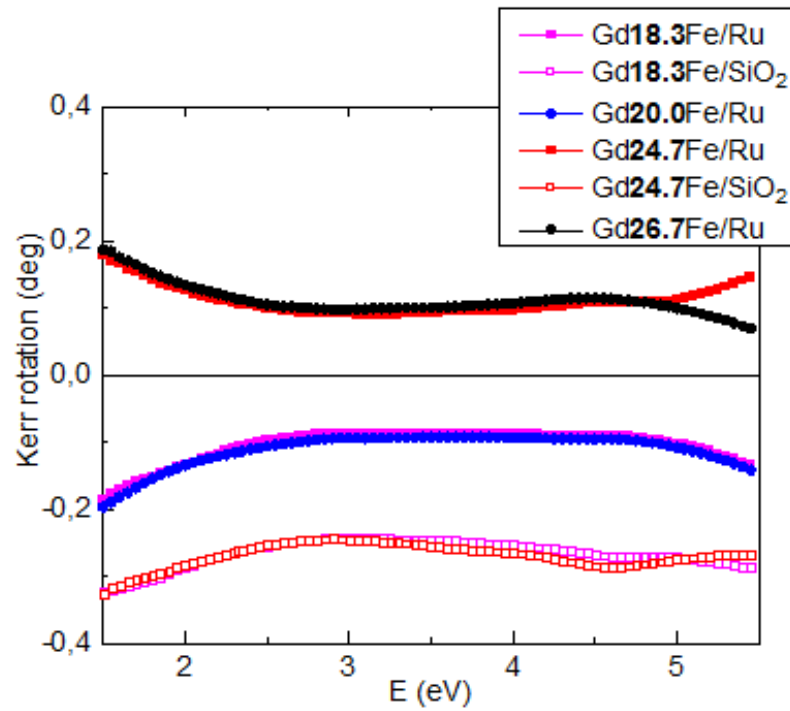


Figure 6.1.8: Polar MOKE rotation spectra of examined  $Gd_xFe_{(100-x)}$  samples with Ru and  $SiO_2$  coatings.

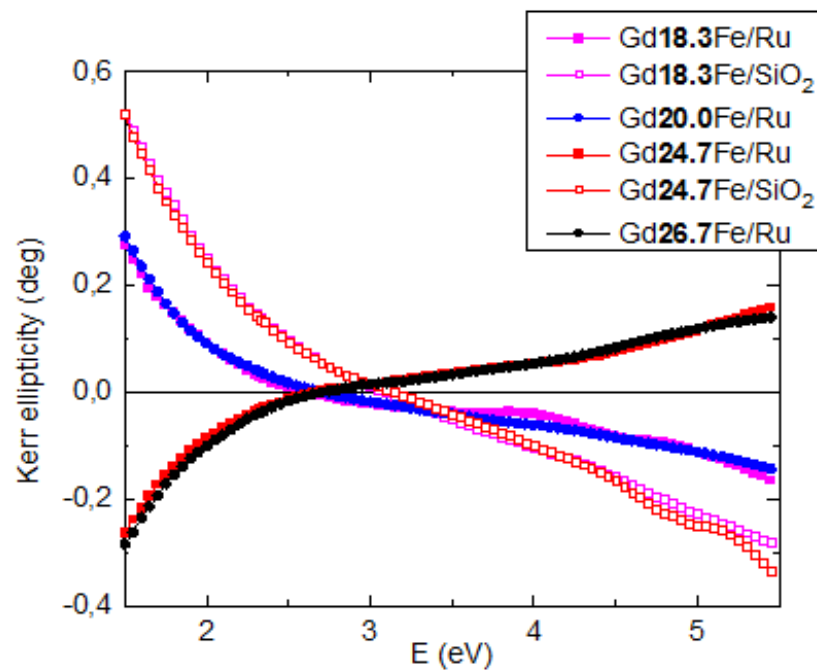


Figure 6.1.9: Polar MOKE ellipticity spectra of examined  $Gd_xFe_{(100-x)}$  samples with Ru and  $SiO_2$  coatings.

We used obtained MOKE spectra to calculate the off-diagonal elements of the  $Gd_xFe_{(100-x)}$  permittivity tensors. For the off-diagonal elements calculations, we used the diagonal elements of the permittivity tensors and thicknesses determined by SE. Figure 6.1.10 shows calculated real parts of the off-diagonal elements  $\epsilon_{2r}$ . Figure 6.1.11 shows calculated imaginary parts of the off-diagonal elements  $\epsilon_{2i}$ . Firstly, the  $\epsilon_{2r}$  spectra are characteristic by one global extreme around 2.5 eV while  $\epsilon_{2i}$  amplitudes decrease with energy in the measured spectral range. Amplitudes of  $Gd_xFe_{(100-x)}$  off-diagonal permittivity elements spectra are smaller than amplitudes of Fe. This is the most probably caused by the presence of the Gd. To discuss the effect of Gd, it is also important to note, that Gd substitution decreases amplitudes of both, the real and imaginary part of the permittivity tensors. We attributed this to the fact that the magnetic moment of Fe is in this ferrimagnetic alloy stronger than the magnetic moment of Gd. Furthermore, change in the sign around the compensation point confirms change of the direction of the net magnetic moment in investigated samples. Apart from the sign change, the spectral behavior remains the same for all samples (it is clearly demonstrated by the same zero crossing near 1.8 eV in the Figure 6.1.10. This indicates only negligible changes in the electronic structure of  $Gd_xFe_{(100-x)}$  with composition around the compensation point. The results are consistent with hysteresis loop measurements in Figure 6.1.7.

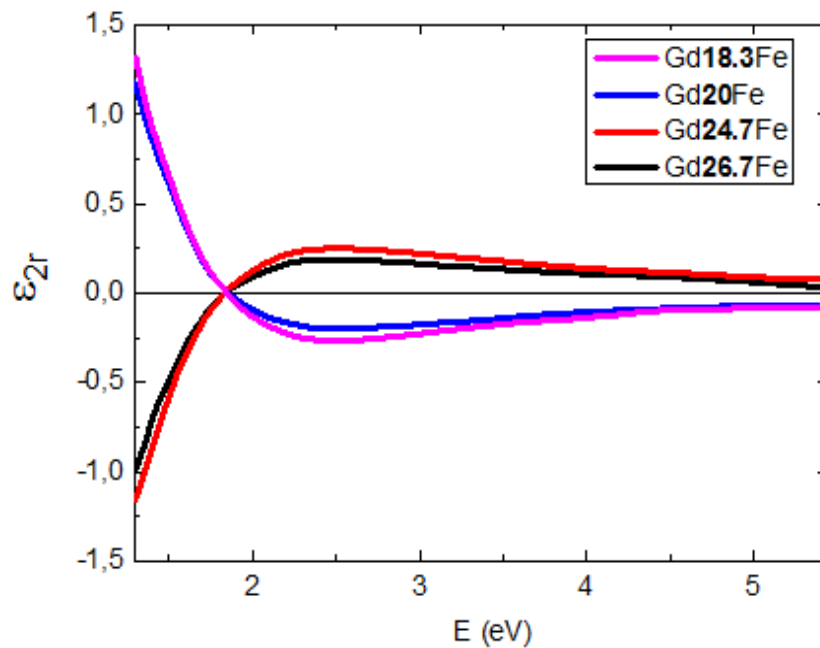
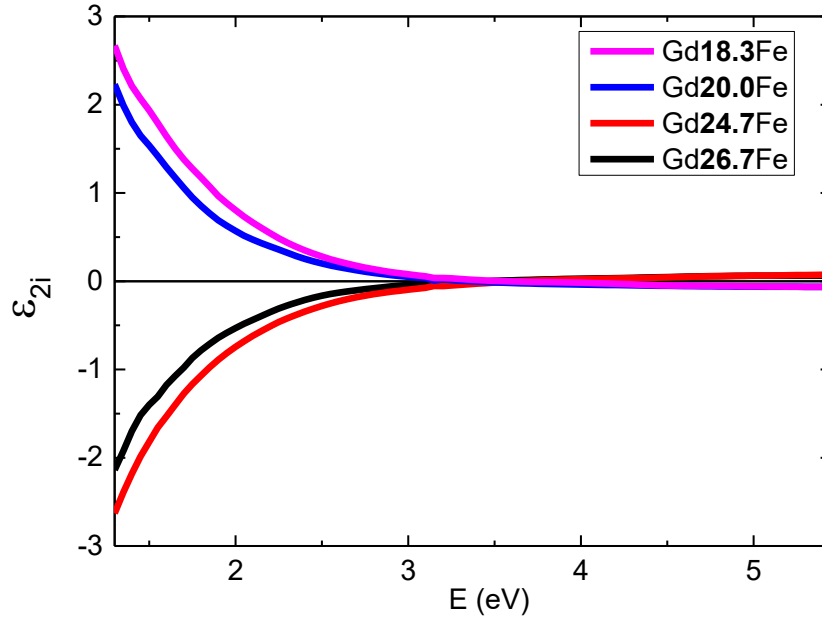


Figure 6.1.10: Real parts of the off-diagonal elements of the permittivity tensors of  $Gd_xFe_{(100-x)}$ .



**Figure 6.1.11: Imaginary parts of the off-diagonal elements of the permittivity tensors of  $Gd_xFe_{(100-x)}$ .**

We parameterized spectra of the off-diagonal elements of the  $Gd_xFe_{(100-x)}$  permittivity tensor in terms of microscopic theory. For this purpose, we used one Dia transition term for all the compositions. Its parameters are listed in table 6.1.3. As one can see, increased Gd concentration is increasing amplitude of the transition only. From these results one can assume that the MO effect observed in MOKE spectra comes from different probabilities of transition between an orbital singlet ground state and excited state split by the combined effect of spin-orbit coupling and an exchange field and that Gd concentration is decreasing this splitting.

**Table 6.1.3: Fitted parameters of Dia and Para transitions used to parameterize off-diagonal elements of the  $Gd_xFe_{(100-x)}$  permittivity tensor in the spectral range from 1.5 to 5.5 eV. Here,  $E_0$  stands for central energy of the transition;  $Amp$  represents an amplitude of the transition and  $\Gamma_0$  is a half-width in the half-height of the transition.**

	Dia tr. 1		
	$E_0$ (eV)	$Amp$	$\Gamma_0$ (eV)
Gd18.3Fe	1	1.7	0.8
Gd20.0Fe	1	1.9	0.8
Gd24.7Fe	1	2.2	0.8
Gd26.7Fe	1	2.4	0.8



## 6.2. Magnetic garnets

Magnetic garnets are crystalline materials with a common structure shown in Figure 6.2.1 and described by a general formula  $\{A^{3+}\}_3[B^{3+}]_2(C^{3+})_3O_{12}$ . In this formula, A stands for doubly positively charged metal ion (Y, Nd, Yb, Lu) surrounded by 8 oxygen ions. B stands for metal ions with three or four positive charges (Fe, Bi, Ga, Al), surrounded by 6 oxygen ions and C for mostly ions with 4 positive charges (Fe, Bi, Ga, Al) surrounded by 4 oxygen ions [29].

These complex materials have recently attracted a considerable attention as they have high application potential. This is mainly given by several magneto-electric, spintronic and MO phenomena, such as spin Seebeck effect [50], spin Hall magneto-resistance [51] as well as high MOKE and Faraday effect in the visible-light region. In this sub-chapter we focus on bismuth substituted yttrium iron garnets  $Y_{3-x}Bi_xFe_5O_{12}$  (Bix:YIGs); as well as on bismuth and gallium substituted neodymium iron garnets  $Nd_2BiFe_{(5-x)}Ga_xO_{12}$  (Bi1:NIGxGs) and  $Nd_{0.5}Bi_{2.5}Fe_{(5-x)}Ga_xO_{12}$  (Bi2.5:NIGxGs). Both materials exhibit strong spin-orbit coupling enhanced by 6p orbitals of Bi. Furthermore, Bi1:NIGxGs and Bi2.5:NIGxGs exhibit strong out-of-plane magnetic anisotropy achieved by Ga substitution and crystal orientation (111) of the GGG substrate. Currently, people use various techniques to grow magnetic garnet thin films of high optical and MO quality [52-65]. As explained in sub-chapter 5.2, MOD has demonstrated to be a very promising method for this type of material.

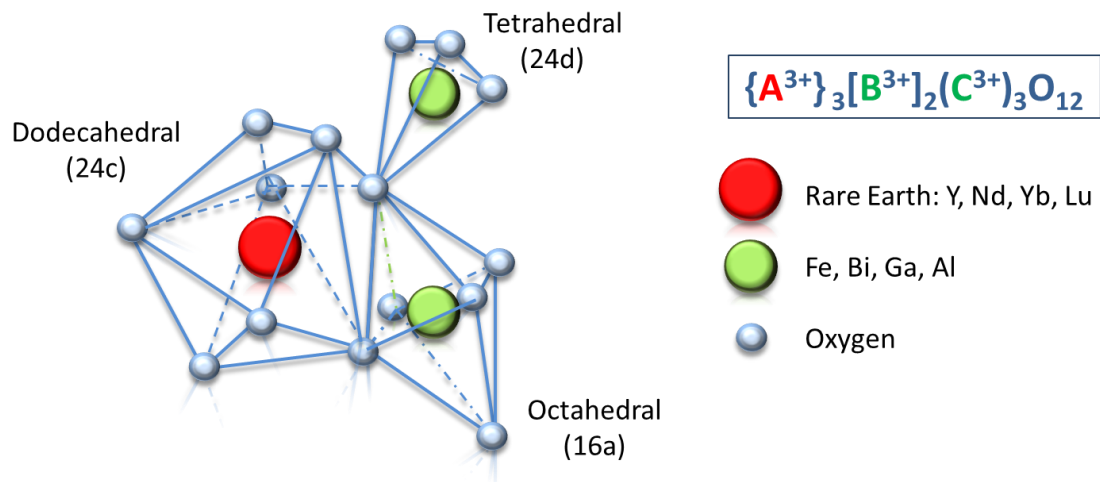
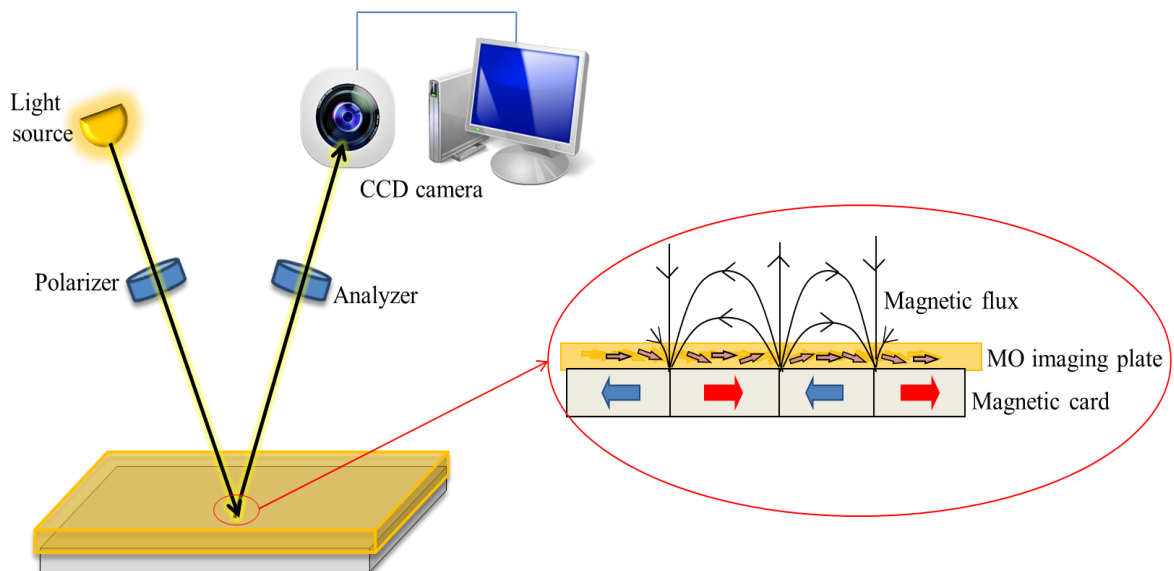


Figure 6.2.1: Atomic structure common to magnetic garnets.

All mentioned properties combined make studied garnets suitable for applications such as magnetic recording or non-reciprocal photonic devices (including optical isolators and circulators). Moreover, garnets are much desired material for various MO visualizers and microscopes [66, 67]. Figure 6.2.2 shows MOKE microscope which enable to visualize magnetic domains on the surface of magnetic materials. In this device, polarized light reflects on the magnetic sample (for example magnetic credit card) and passes through an analyzer (polarizing filter), before going through a regular optical microscope. When the polarized light reflects on the magnetic sample, MOKE causes different changes in the light polarization for differently oriented magnetic domains. These polarization changes are afterwards converted by the analyzer into the light intensity changes, which are visible. However, magnetic domains are not easily to observe for materials that exhibit small MO effects or materials that are covered by a non-transparent protecting layer (plastic layer in cards). Therefore, to make magnetic domains visible, MOKE microscope uses transparent MO imaging plate that exhibits huge MO effect (garnet film). This MO plate is put on the sample and it copies its magnetic field. Therefore, we do not observe MO effect on examined sample, but on MO plate with identical magnetic domains shape as the sample.



**Figure 6.2.2: MOKE microscope principle and magnetic response of MO imaging plate to a material with magnetic domains.**

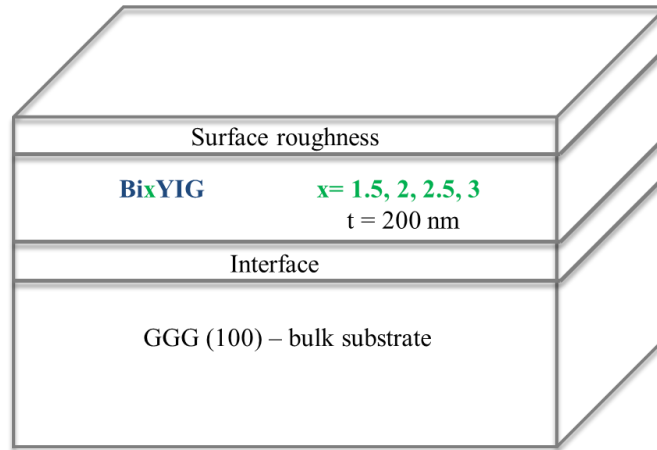
### 6.2.1 $Y_{3-x}Bi_xFe_5O_{12}$

In this sub-chapter, we focused on the determination of complete permittivity tensors of Bix:YIGs thin films with various Bi concentrations. We used optical and MO spectral measurements at energies from 1.5 to 5 eV. We compared obtained results to the properties of investigated Bix:YIGs thin films prepared by MOD to properties of bulk-like Bix:YIGs with small Bi concentrations prepared by epitaxial growth [8, 68]. We examined optical properties by SE supported by transmission intensity measurements. From these data we derived the diagonal permittivity tensor elements. We examined MO properties by spectroscopic MOKE and Faraday effect measurements and analysis. Using a combination of the SE and MO measurements we determined the spectral dependence of off-diagonal permittivity tensor elements. Finally, we parameterized obtained results in terms of microscopic theory which relates permittivity tensor spectra to the energy-level splitting and transition probabilities.

We focused on  $Y_{3-x}Bi_xFe_5O_{12}$  thin films ( $x= 1.5, 2, 2.5, 3$ ) prepared on  $Gd_3Ga_5O_{12}$  (GGG) (100) substrates. We listed compositions of garnet films in the table 6.2.1. The thin films were prepared by MOD method. MOD liquids for garnet films consisted of solutions made of Bi, Y, and Fe carboxylates. The total concentration of carboxylates was 3 – 4% [20, 69]. We prepared MOD liquids by mixing each solution to obtain desired chemical compositions. We spin-coated MOD liquids on GGG(100) substrates using 3000 rpm for 60 s. We followed this process by drying at 100 °C for 30 minutes using a hot plate. Afterwards, we pre-annealed samples at 450 °C for 30 minutes to decompose organic materials and obtain amorphous oxide films. We repeated procedure from spin coating to pre-annealing four or five times to obtain appropriate thicknesses. Nominal thicknesses of studied garnet films were 160 and 200 nm (see table 6.2.1). We determined nominal thicknesses from the number of MOD cycles calibrated by X-ray measurements. Finally, we annealed samples for the crystallization in a furnace using 700 °C for 3 hours. We performed all thermal treatments in the air. For further information on garnet films prepared by MOD see Ref. [19]. Figure 6.2.3 shows theoretical model structure of Bix:YIGs samples used for SE, MOKE and Faraday effect analyses.

**Table 6.2.1: Compositions and nominal thicknesses of examined garnet films.**

Film composition	Indication in Figures and text	Nominal thickness (nm)
$\text{Bi}_3\text{Fe}_5\text{O}_{12}$	Bi3IG	200
$\text{Y}_{0.5}\text{Bi}_{2.5}\text{Fe}_5\text{O}_{12}$	Bi2.5YIG	160
$\text{Y}_1\text{Bi}_2\text{Fe}_5\text{O}_{12}$	Bi2YIG	200
$\text{Y}_{1.5}\text{Bi}_{1.5}\text{Fe}_5\text{O}_{12}$	Bi1.5YIG	200

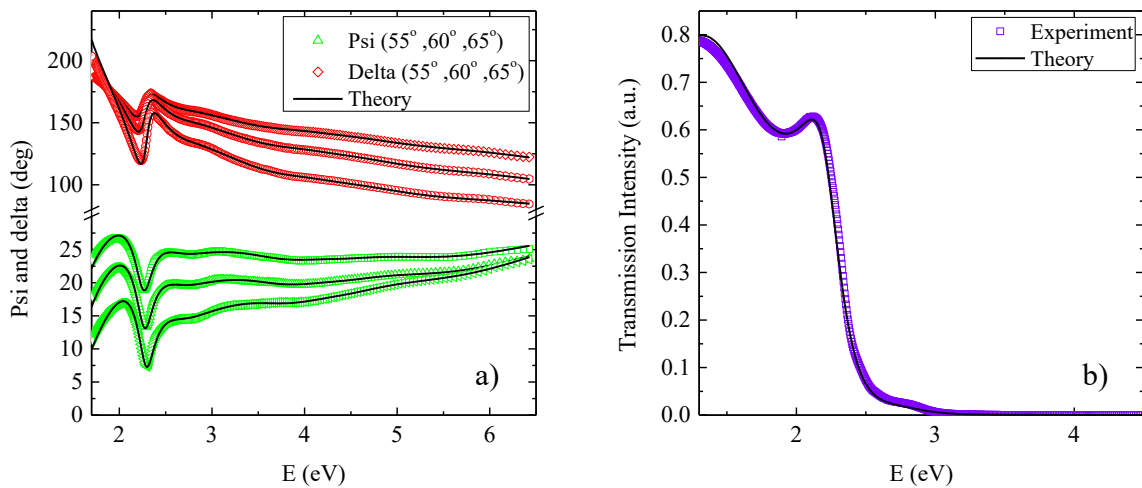


**Figure 6.2.3: Model structure of Bix:YIGs samples used for SE, MOKE and Faraday analysis calculations.**

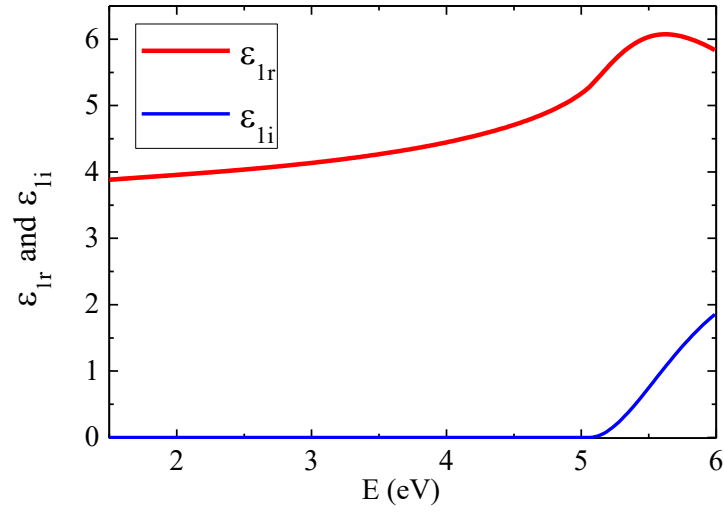
### a) Spectroscopic Ellipsometry

We performed SE measurements on a Mueller matrix ellipsometer Woollam RC2. We measured spectral dependence of ellipsometry parameters  $\psi$  and  $\Delta$  in reflection and at incident angles  $55^\circ$ ,  $60^\circ$  and  $65^\circ$ . We used the same equipment to measure the transmission spectra at the incidence angle  $0^\circ$ . All measurements were performed in the spectral range from 1.5 to 6.5 eV. We analyzed SE experimental data using a CompleteEase software provided by Woollam Co.. We used MSA mode to obtain optical functions spectra (diagonal elements of the permittivity tensor  $\epsilon_{1r}$  and  $\epsilon_{li}$ ) of GGG and Bix:YIGs materials. In MSA mode, we combined ellipsometry and transmission measurements for each material. We used transmission spectra because of the strong interference observed in  $\psi$  and  $\Delta$  in the transparent spectra region below 2.5 eV. We fitted the SE and transmission experimental data using model structure shown in Figure 6.2.3. Figure 6.2.4 shows that theoretical model describes both: SE

and transmission experimental spectra well. To ensure Kramers-Kronig consistent results we parameterized obtained optical functions. We parameterized optical functions of GGG substrate (Figure 6.2.5) by one Tauc-Lorentz oscillator in whole measured spectral range. We parameterized calculated optical functions of Bi<sub>3</sub>YIGs by three Gaussian oscillators (especially in the spectral range 3-5.5 eV) and one general Herzinger-Johns oscillator (especially below 3 eV). Since substrates were transparent and both-side polished, we considered back reflections in the SE analysis. We fitted all thicknesses (including roughness and interface layer) by the CompleteEase software and subsequently used them in MOKE and Faraday spectra analysis. Table 6.2.2 shows some parameters of used oscillators. Table 6.2.3 shows fitted thicknesses.



**Figure 6.2.4: Experimental data for Bi<sub>3</sub>YIG layer on GGG substrate compared to the theoretical model. a) Variable angle Psi and Delta SE data. b) Measured transmission intensity spectra.**



**Figure 6.2.5: Spectral dependence of diagonal elements  $\epsilon_{lr}$  and  $\epsilon_{li}$  of the GGG (100) substrate permittivity tensor.**

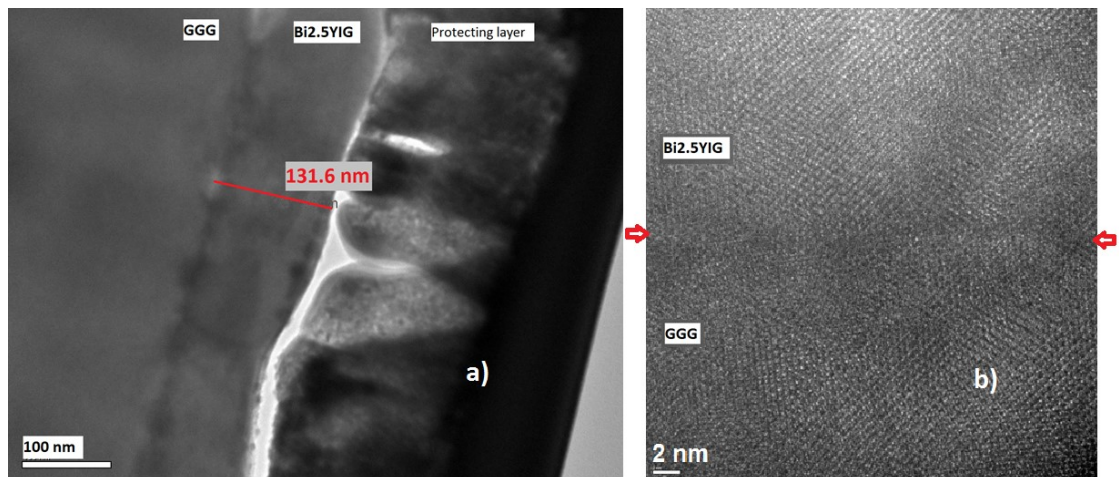
**Table 6.2.2: Fitted parameters of Gaussian and Herzinger-Johns functions used to parameterize optical functions of Bix:YIGs layers on GGG substrates in the spectral range 1.5 - 6.5 eV. Here,  $E_0$  stands for central energy of the function; Amp represents amplitude of the function and Br is its broadening.**

	Gaussian 1			Gaussian 2			Gaussian 3			Herzinger-Johns	
	$E_0$ (eV)	Amp	Br (eV)	$E_0$ (eV)	Amp	Br (eV)	$E_0$ (eV)	Amp	Br (eV)	$E_0$ (eV)	Amp
Bi3IG	3.27	1.64	0.81	4.36	3.4	1.65	6.27	5.05	3.09	2.60	2.33
Bi2.5YIG	3.2	2.7	1.15	4.33	2.29	1.64	6.51	4.26	3.87	2.49	2.40
Bi2YIG	3.04	1.15	0.77	4.25	1.56	1.68	6.36	2.98	2.37	2.53	1.86
Bi1.5YIG	2.71	0.83	0.46	4.23	0.99	2.19	7.31	2.13	4.18	2.89	1.46

**Table 6.2.3: Fitted thicknesses and volume fractions used for model of Bix:YIGs layers on GGG substrate in the spectral range 1.5 - 6.5 eV. Here,  $T_{\text{Bi:YIG}}$  stands for Bi:YIG film thickness;  $R_{\text{rough}}$  represents film roughness with volume fractions  $f_{\text{rough}}$ ;  $R_{\text{interf}}$  represents thickness of film/GGG interface with volume fractions  $f_{\text{interf}}$ .**

	$T_{\text{Bi:YIG}}$ (nm)	$R_{\text{rough}}$ (nm)	$f_{\text{rough}}$	$R_{\text{interf}}$ (nm)	$f_{\text{interf}}$
Bi3IG	177	7	0.5	4	0.5
Bi2.5YIG	132	5	0.4	2	0.5
Bi2YIG	165	12	0.6	3	0.5
Bi1.5YIG	175	12	0.5	4	0.5

We performed transmission electron microscopy (TEM) measurement on Bi2.5YIG sample. We did this measurement to observe the film quality and to confirm thicknesses derived from SE. Figure 6.2.6 shows results of the TEM measurement. This measurement confirmed that the film was uniform and epitaxially grown on GGG. (This was expected since X-ray diffraction data of previously grown Bix:YIGs in our laboratory also revealed garnet structure [70].) TEM also revealed that the film thickness was around 132 nm, which is almost 30 nm less than the nominal thickness. Moreover, we can observe interfacial layer in the form of contrast layer between GGG and Bi2.5YIG. These observations are in a particularly good agreement with the SE analysis results (Table 6.2.3). Finally, we performed AFM roughness measurement on Bi1.5YIG sample. We did this measurement to verify relatively high roughness derived from SE. This measurement revealed roughness 11 nm which is in a good agreement with SE result.



**Figure 6.2.6: a) TEM picture of the Bi2.5YIG sample. b) TEM picture of the interface between GGG and Bi2.5YIG layer.**

Figures 6.2.7 and 6.2.8 shows spectra of calculated optical functions  $\epsilon_{lr}$  and  $\epsilon_{li}$  for all Bix:YIG samples. In case of  $\epsilon_{lr}$  we observed one global maximum around 2.4 eV and two local maxima around 3.2 and 4.4 eV. We also observed optical transitions around 2.5, 3.2 and 4.4 eV. The absorption edge near 2.1 eV was clearly visible. Bi3YIG and Bi2.5YIG spectra have in the UV region similar shape. This shape is however different from Bi2YIG and Bi1.5YIG spectra. This observation corresponds to the off-diagonal elements of the permittivity tensor spectra discussed later in this

sub-chapter, where we observe a bigger change in absorption between Bi2YIG and Bi2.5YIG samples. All spectra clearly demonstrated that the bismuth substitution increases amplitudes of  $\epsilon_{1r}$  and  $\epsilon_{1i}$  in the measured spectral range.

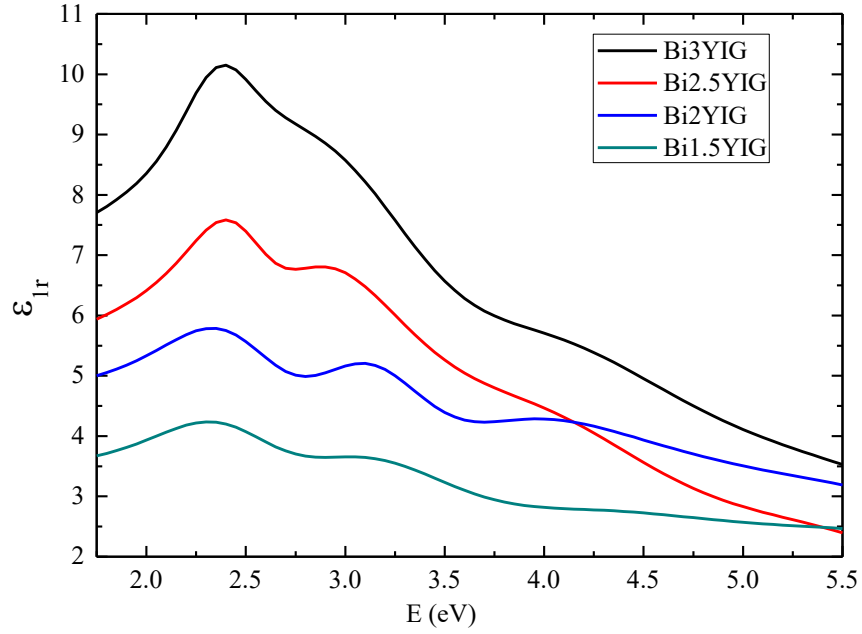


Figure 6.2.7: Real parts of diagonal elements of the permittivity tensor of Bix:YIGs.

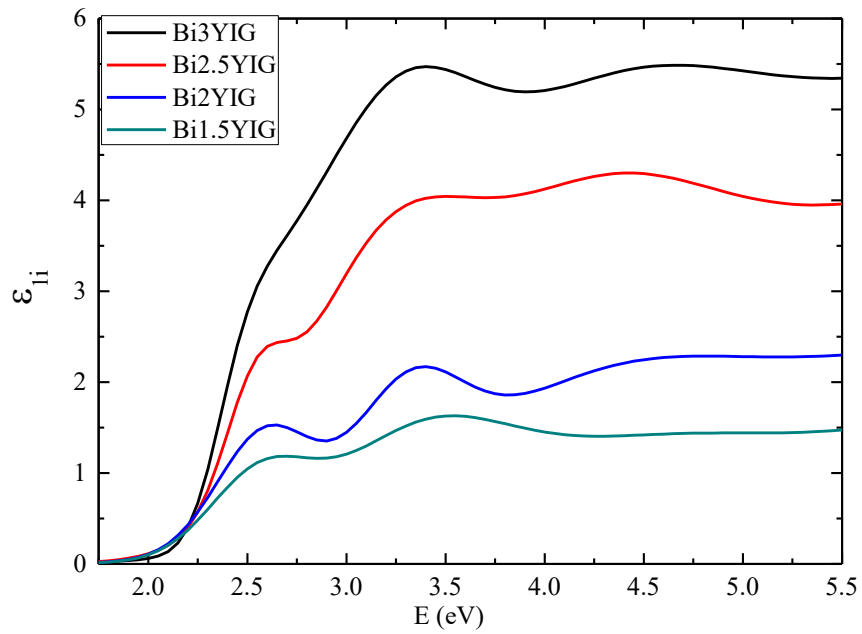


Figure 6.2.8: Imaginary parts of diagonal elements of the permittivity tensor of Bix:YIGs.



When we compare our results to the results previously reported on bulk-like films [8], we can notice smaller  $\epsilon_{1i}$  amplitude and an absorption edge shift from 2.5 to 2.2 eV. However, similar measurements performed on thin single crystal Bix:YIGs films show absorption data similar to our results [71-73]. Therefore we attributed this discrepancy to the difference between properties of thin and bulk-like films. Previous investigations [74-78] demonstrated that properties of ultra-thin films may significantly differ from properties of thicker films. Difference is usually caused either by materials' inhomogeneities or, as the thickness of the films decreases, by the increasing influence of surface and interface defective layers (in here modeled by EMA) [76, 77, 79].

#### **b) MOKE and Faraday effect spectroscopy**

We measured Bix:YIGs MO properties by MOKE and MO Faraday effect spectroscopy. We measured MOKE rotation and ellipticity spectra in the polar configuration. We acquired the spectra at room temperature and at nearly normal light incidence. Applied magnetic field was 1.2 T which was enough for samples saturation. We used p-polarized light. We recorded data in the photon energy range from 1.4 to 5 eV. Faraday rotation and ellipticity spectra were acquired at room temperature using magnetic field 665 mT, which was enough for samples saturation. We recorded Faraday experimental spectra in the photon energy range from 1.4 to 4 eV. Faraday hysteresis loops were measured at 3 eV.

Figure 6.2.9 shows measured MOKE rotation spectra. Figure 6.2.10 shows measured MOKE ellipticity spectra. We observed MOKE rotation maxima around 3.4 and 4 eV and MOKE ellipticity maxima near 3.3 and 4.4 eV. These values are characteristic for Bix:YIGs MOKE spectra [8, 68]. Furthermore, we observed strong MO interference in the form of strong oscillations in the spectral range below 3 eV. Since our samples had different thicknesses, we observed different interference patterns for each of them. Spectra clearly demonstrated that bismuth substitution increases amplitudes of MOKE rotation and ellipticity effectively in the measured spectral range.

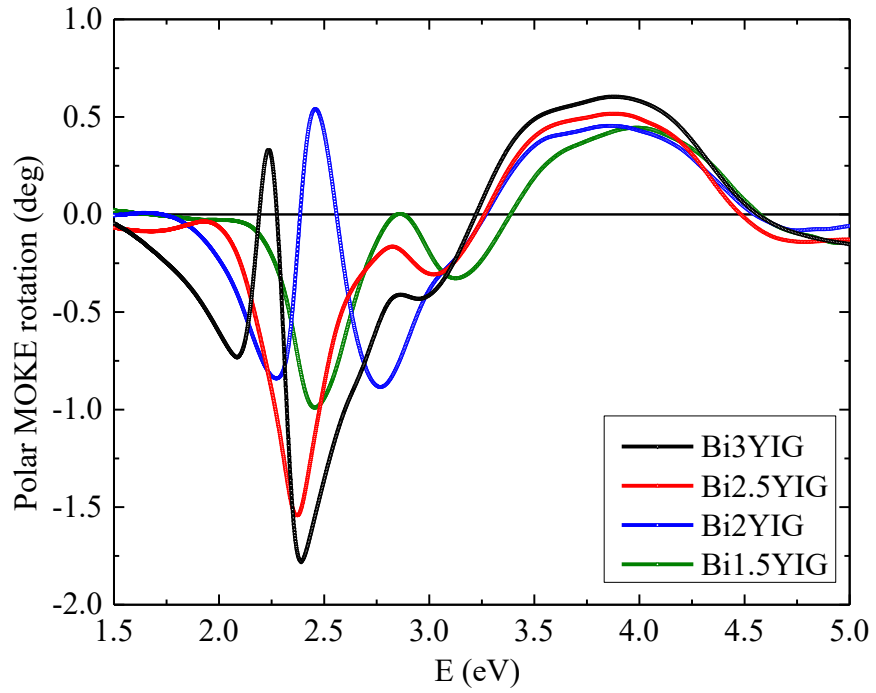


Figure 6.2.9: Polar MOKE rotation spectra of Bi<sub>x</sub>:YIGs on GGG substrates.

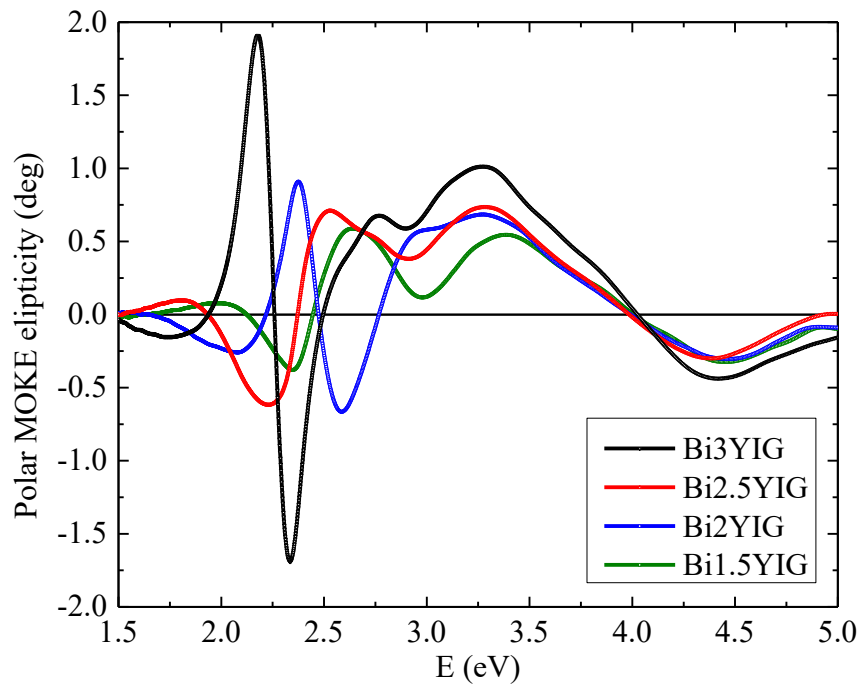


Figure 6.2.10: Polar MOKE ellipticity spectra of Bi<sub>x</sub>:YIGs on GGG substrates.

Figure 6.2.11 shows measured MO Faraday rotation spectra. Figure 6.2.12 shows measured MO Faraday ellipticity spectra. Experimental data were corrected for the rotation from the substrate. We observed Faraday rotation minima near 2.4 eV and two maxima near 2.7 and 3.2 eV. Faraday ellipticity showed maxima at 2.7 and minima at 3.3 eV. As expected, spectra demonstrated that bismuth substitution leads to the enhancement of the MO Faraday rotation near to 2.4, 2.7 and 3.2 eV and ellipticity near 2.7 eV. We demonstrated Faraday rotation angle enhancement by Faraday hysteresis loop measurements shown in Figure 6.2.13.

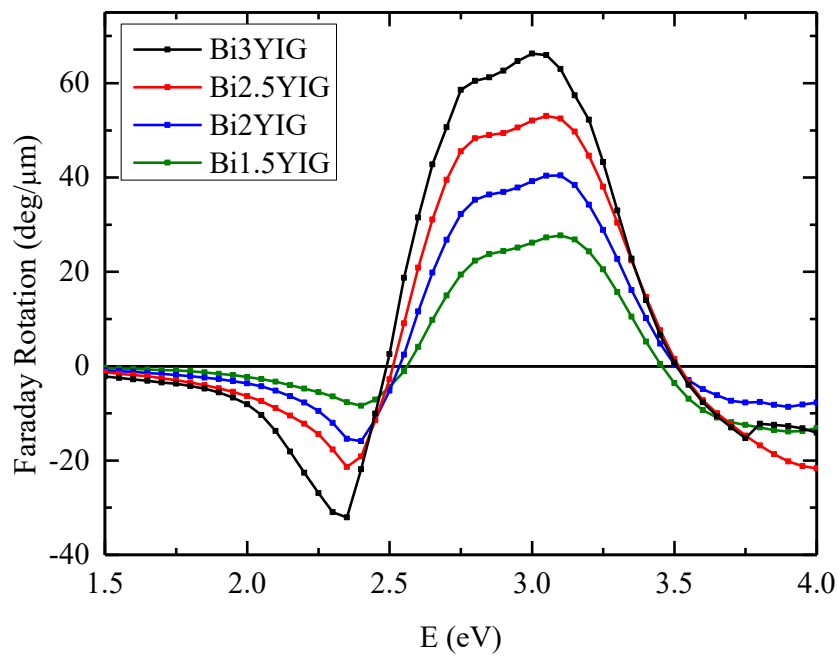


Figure 6.2.11: Faraday effect rotation spectra of Bix:YIGs on GGG substrates.

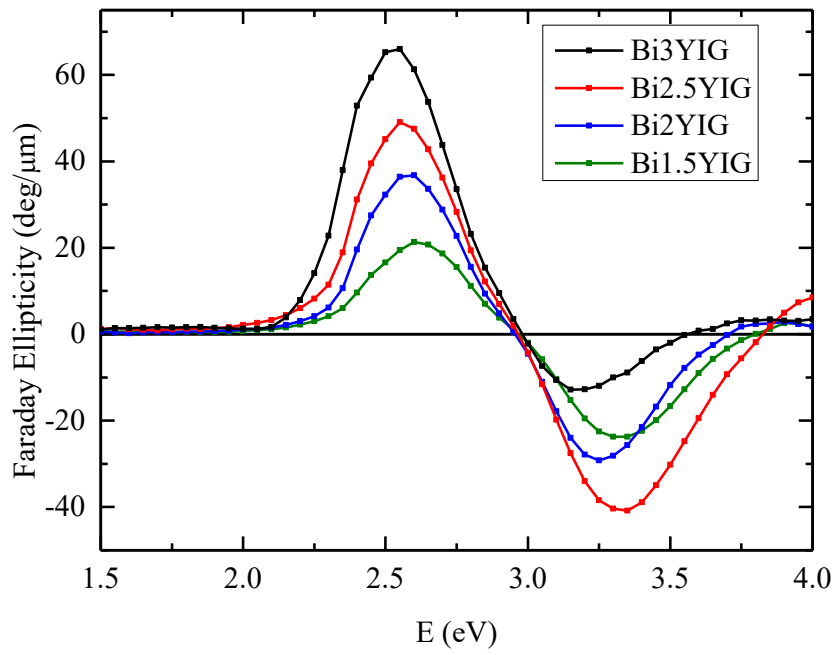


Figure 6.2.12: Faraday effect ellipticity spectra of Bi<sub>x</sub>:YIGs on GGG substrates.

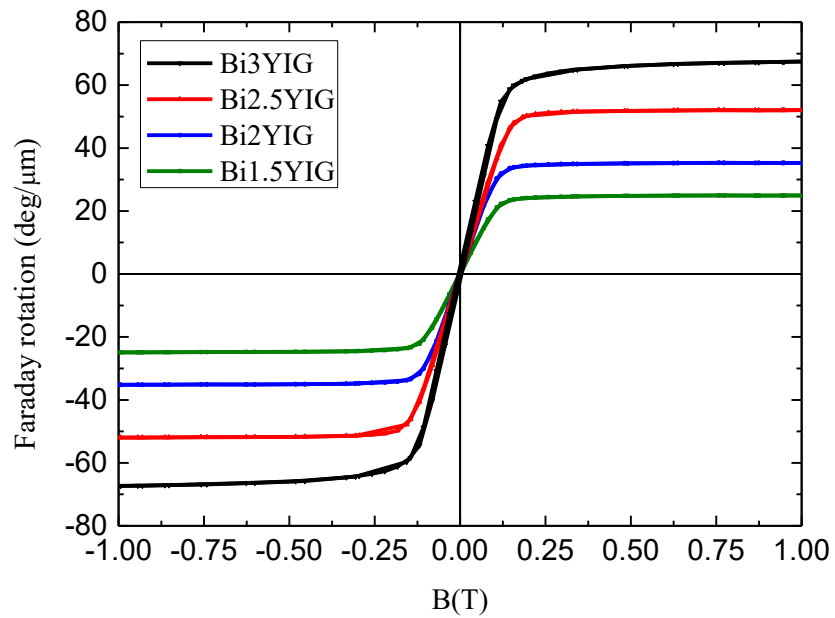


Figure 6.2.13: Faraday effect rotation hysteresis loops of Bi<sub>x</sub>:YIGs on GGG substrates.

We calculated the off-diagonal elements of the permittivity tensor  $\epsilon_{2r}$  and  $\epsilon_{2i}$  from MOKE and Faraday effect spectra. We performed calculations using Yeh's 4x4 matrix formalism and diagonal elements of the permittivity tensor determined by SE. As mentioned previously, there was strong MO interference below 3 eV in MOKE spectra. Therefore, we used MO Faraday effect spectra in the spectral range from 1.5 to 3 eV. Accordingly, we used MOKE spectra in the spectral range from 3 to 5 eV. Figure 6.2.14 shows the real part of off-diagonal permittivity tensor elements  $\epsilon_{2r}$  spectra. We observed sharp global maxima at 2.4 eV and local maxima at 4.2 eV. Minima around 2.7 eV and 3.1 eV were also observed, especially for higher Bi substitutions. As expected, bismuth increases  $\epsilon_{2r}$  amplitudes at extremes 2.4, 2.7 and 3.1 eV significantly.

Figure 6.2.15 shows the imaginary part of off-diagonal permittivity tensor elements  $\epsilon_{2i}$  spectra. We observed clear maxima at 2.5 and 4.5 eV and one minimum near 3.4 eV. Spectra demonstrated that bismuth substitution increases amplitudes of  $\epsilon_{2i}$ , especially at extremes 2.3, 3.4 and 4.5 eV.

We parameterized spectra of the off-diagonal elements of the Bix:YIGs permittivity tensors  $\epsilon_{2r}$  and  $\epsilon_{2i}$  in terms of microscopic theory. For this purpose, we used two Para transition terms at 2.4 and 3.1 eV and three Dia transition terms at 0.5, 2.5, 3.3 and 4.45-4.65 eV. We used the Dia transition at 0.5 eV only to model the effect of the transitions outside of the measured spectral range. Therefore, we do not attribute any physical meaning to it. We listed some of used parameters in table 6.2.4. From these data, we can see that Bi substitution increases amplitudes of almost all listed transitions. Moreover, it lowers energy of Dia transition at 4.65 eV which most likely exist due to charge transfers from oxygen to octahedral Fe [8]. We associated Dia transitions at 2.5 eV and 3.3 eV, which are the strongest, with transitions  $t_2(\text{Fe}^{3+}) \rightarrow t_2g(\text{Fe}^{2+})$  and  $eg(\text{Fe}^{3+}) \rightarrow e(\text{Fe}^{2+})$ . These transitions are mainly responsible for the remarkable increase of the Faraday rotation in the visible and near infrared region. In here we note that in studied materials, Bi is substituted per Y and not for the Fe. Therefore, the positive impact of Bi on mentioned transitions have been previously explained by increase in super-exchange interaction caused by an enhancement of electronic exchange [80]. This enhancement is the most probably facilitated by mixing 6p orbitals of Bi with 2p orbitals of Oxygen and 3d orbitals of iron. This

leads to considering of Bi ion as a magnetic ion [8, 80, 81]. Finally, maxima at 2.4 eV are mainly created by the overlap of secondary negative peaks of these two dominant Dia transitions [8].

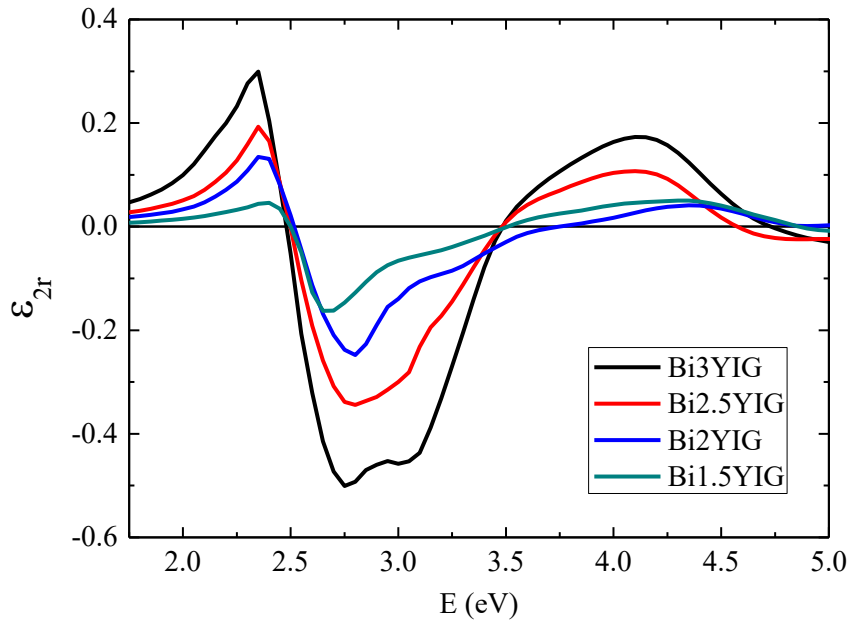


Figure 6.2.14: Real parts of off-diagonal elements of the permittivity tensor of Bi<sub>x</sub>:YIGs.

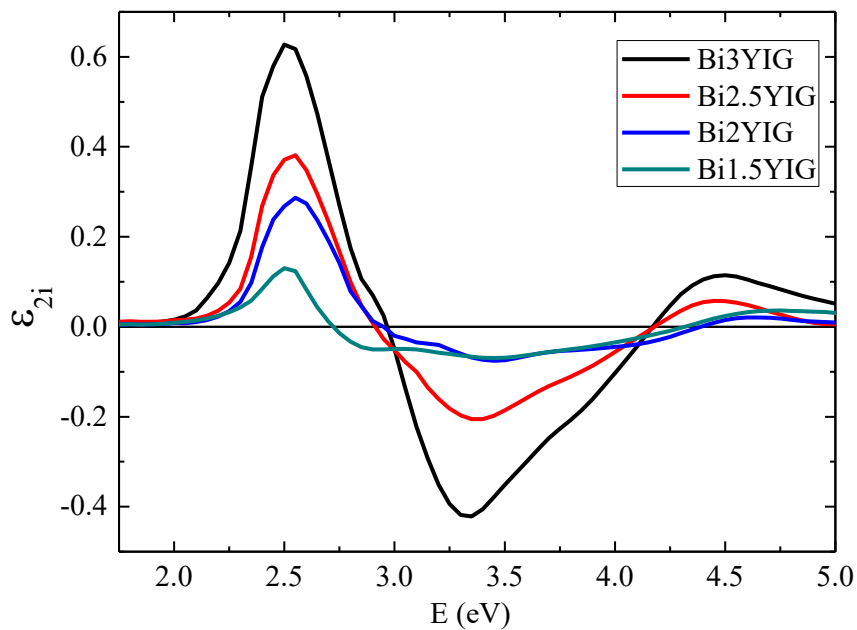


Figure 6.2.15: Imaginary parts of off-diagonal elements of the permittivity tensor of Bi<sub>x</sub>:YIGs.

**Table 6.2.4: Fitted parameters of Dia and Para transitions used to parameterize off-diagonal elements of Bix:YIGs permittivity tensors in the spectral range from 1.5 to 5.5 eV. Here,  $E_0$  stands for central energy of the transition;  $Amp$  represents amplitude of the transition and  $\Gamma_0$  is half-width in a half-height of the transition.**

	Dia tr. 1			Dia tr. 2			Dia tr. 3		
sign	1			1			-1		
	$E_0$ (eV)	Amp	$\Gamma_0$ (eV)	$E_0$ (eV)	Amp	$\Gamma_0$ (eV)	$E_0$ (eV)	Amp	$\Gamma_0$ (eV)
Bi3IG	0.5	0.03	0.4	2.5	0.8	0.17	3.3	0.5	0.4
Bi2.5YIG	0.5	0.03	0.4	2.55	0.5	0.2	3.3	0.25	0.4
Bi2YIG	0.5	0.03	0.4	2.55	0.35	0.2	3.3	0.1	1
Bi1.5YIG	0.5	0.03	0.4	2.55	0.2	0.15	3.3	0.1	1

Dia tr. 4			Para tr. 1			Para tr. 1		
1			1			-1		
$E_0$ (eV)	Amp	$\Gamma_0$ (eV)	$E_0$ (eV)	Amp	$\Gamma_0$ (eV)	$E_0$ (eV)	Amp	$\Gamma_0$ (eV)
4.45	0.15	0.3	2.4	0.09	0.2	3.1	0.05	0.15
4.5	0.1	0.3	2.45	0.08	0.2	3.1	0	0.15
4.55	0.05	0.4	2.45	0.06	0.2	3.1	0	0.15
4.65	0.06	0.55	2.45	0.02	0.2	3	0.01	0.15

We would like to note in here that all calculated permittivity tensor elements spectra have characteristic shape of diagonal and off-diagonal permittivity tensor elements of bulk-like epitaxial Bix:YIGs with small Bi concentrations [8, 68]. As explained earlier, properties of epitaxial films may differ from the properties of bulk-like materials. Therefore, we attributed result discrepancy to the fact that in this work we characterized epitaxial thin films. Results demonstrated that MOD in as an effective technique for preparation of epitaxial thin garnet films on GGG substrate.

### 6.2.2. $\text{Nd}_2\text{BiFe}_{(5-x)}\text{Ga}_x\text{O}_{12}$ & $\text{Nd}_{0.5}\text{Bi}_{2.5}\text{Fe}_{(5-x)}\text{Ga}_x\text{O}_{12}$

In this sub-chapter, we focused on the determination of complete permittivity tensors of Bi1:NIGxGs and Bi2.5:NIGxGs thin films. We used optical and MO spectral measurements at energies from 1.5 to 5.5 eV. We examined optical properties by SE supported by transmission intensity measurements. From these data we derived the

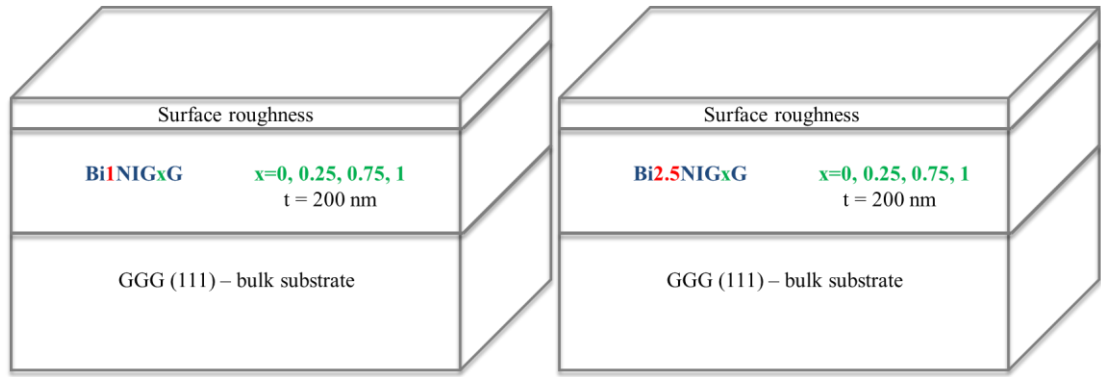
diagonal permittivity tensor elements. We examined MO properties by spectroscopic polar MOKE and Faraday effect rotation and ellipticity measurements. From these data we determined the spectral dependence of off-diagonal permittivity tensor elements. Finally, we parameterized obtained results in terms of microscopic theory.

We focused on  $\text{Nd}_2\text{BiFe}_{(5-x)}\text{Ga}_x\text{O}_{12}$  and  $\text{Nd}_{0.5}\text{Bi}_{2.5}\text{Fe}_{(5-x)}\text{Ga}_x\text{O}_{12}$  thin films ( $x = 0, 0.25, 0.75, 1$ ) prepared by MOD method on  $\text{Gd}_3\text{Ga}_5\text{O}_{12}$  (GGG) (111) substrates. Compositions of garnet films are listed in the table 6.2.5. MOD liquids for garnet films consisted of solutions made of Nd, Bi, Ga, and Fe carboxylates [20, 69]. We prepared MOD liquids by mixing each solution to obtain desired chemical compositions. We spin-coated those solutions on GGG (111) substrates with 3000 rpm for 30 s and continued by drying at 100 °C for 10 minutes using a hot-plate. To decompose organic materials and obtain amorphous oxide films, we pre-annealed samples at 450 °C for 10 minutes. We repeated this procedure, from spin coating to pre-annealing, 5 times to obtain appropriate thickness. Nominal thicknesses of studied garnet films were 200 nm (based on the number of MOD cycles calibrated by X-ray measurements). Finally, we annealed samples for crystallization in a furnace at 700 °C for 3 hours. We performed all the thermal treatments in the air. Figure 6.2.16 shows theoretical model structure of Bi1:NIGxGs and Bi2.5:NIGxGs samples used for SE, MOKE and Faraday effect analysis.

**Table 6.2.5. Compositions and nominal thicknesses of examined garnet films**

Film composition	Indication in Figures and text	Nominal thickness (nm)
$\text{Nd}_2\text{BiFe}_5\text{O}_{12}$	Bi1NIG	200
$\text{Nd}_2\text{BiFe}_{4.75}\text{Ga}_{0.25}\text{O}_{12}$	Bi1NIG(0.25)G	200
$\text{Nd}_2\text{BiFe}_{4.25}\text{Ga}_{0.75}\text{O}_{12}$	Bi1NIG(0.75)G	200
$\text{Nd}_2\text{BiFe}_4\text{GaO}_{12}$	Bi1NIGG	200
$\text{Nd}_{0.5}\text{Bi}_{2.5}\text{Fe}_4\text{GaO}_{12}$	Bi2.5NIG	200
$\text{Nd}_{0.5}\text{Bi}_{2.5}\text{Fe}_{4.75}\text{Ga}_{0.25}\text{O}_{12}$	Bi2.5NIG(0.25)G	200
$\text{Nd}_{0.5}\text{Bi}_{2.5}\text{Fe}_{4.25}\text{Ga}_{0.75}\text{O}_{12}$	Bi2.5NIG(0.75)G	200
$\text{Nd}_{0.5}\text{Bi}_{2.5}\text{Fe}_4\text{Ga}_1\text{O}_{12}$	Bi2.5NIGG	200





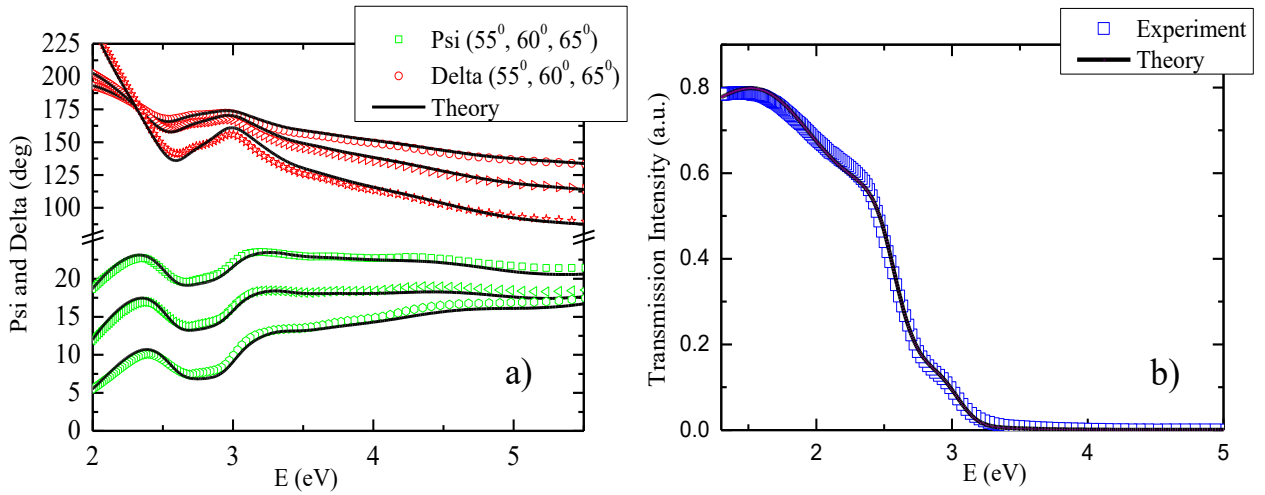
**Figure 6.2.16: Model structure of Bi1NIGG and Bi2.5NIGG samples used for SE, MOKE and Faraday analysis calculations.**

### a) Spectroscopic Ellipsometry

We performed SE measurements on a Mueller matrix ellipsometer Woollam RC2. We measured spectral dependence of ellipsometry parameters  $\psi$  and  $\Delta$  in reflection and at incident angles  $55^\circ$ ,  $60^\circ$  and  $65^\circ$ . We used the same equipment to measure the transmission spectra at the incidence angle  $0^\circ$ . We performed measurements in the spectral range from 1.5 to 6.5 eV.

We analyzed SE experimental data using a CompleteEase software provided by Woollam Co.. We used MSA mode to obtain optical functions spectra (diagonal elements of the permittivity tensor  $\varepsilon_{1r}$  and  $\varepsilon_{1i}$ ) of Bi1:NIGxGs and Bi2.5:NIGxGs. In MSA mode, we combined ellipsometry and transmission measurements of each sample. Transmission spectra supplemented our analysis because of the strong interference observed in  $\psi$  and  $\Delta$  in the transparent region below 2.5 eV. We fitted SE and transmission experimental data using a model structure shown in Figure 6.2.16. To ensure Kramers-Kronig consistent results we parameterized obtained optical functions  $\varepsilon_{1r}$  and  $\varepsilon_{1i}$  of Bi1:NIGxGs and Bi2.5:NIGxGs by five Gaussian functions. Since substrates were transparent and both-side polished, we considered back reflections in the SE analysis. We fitted all thicknesses, including roughness in the CompleteEase software. We subsequently used these thicknesses in MOKE and Faraday spectra analysis. We listed some parameters of used parameterization functions in the Table 6.2.6 and the fitted thicknesses in the Table 6.2.7. Fitted interface thickness was zero for all samples. This zero thickness corresponds to

epitaxial growth of our films on GGG substrate. Figure 6.2.17 shows that used theoretical model describes both: SE and transmission experimental spectra well.



**Figure 6.2.17: Experimental data for Bi1NIG layer on GGG substrate compared to the theoretical model. a) Variable angle Psi and Delta SE data. b) Measured transmission intensity spectra.**

**Table 6.2.6: Fitted parameters of Gaussian functions used to parameterize optical properties of Bi1:NIGxGs and Bi2.5:NIGxGs layers on GGG substrates in the spectral range from 1.5 to 6.5 eV. Here,  $E_0$  stands for central energy of the function; Amp represents amplitude of the function and Br its broadening.**

	Gaussian 1			Gaussian 2			Gaussian 3		
	$E_0$ (eV)	Amp	Br (eV)	$E_0$ (eV)	Amp	Br (eV)	$E_0$ (eV)	Amp	Br (eV)
Bi1NIG	2.75	0.76	0.39	3.27	0.76	0.32	3.42	1.81	0.80
Bi1NIG(0.25)G	2.79	0.72	0.42	3.25	1.11	0.24	3.45	1.64	0.63
Bi1NIG(0.75)G	2.76	0.39	0.38	3.33	1.08	0.62	4.16	2.70	1.58
Bi1NIGG	2.75	0.19	0.29	3.38	1.32	0.84	4.26	1.07	0.88
Bi2.5NIG	2.59	1.35	0.35	3.33	4.91	1.03	4.12	3.12	0.93
Bi2.5NIG(0.25)G	2.61	1.31	0.34	3.17	3.11	0.80	4.03	3.14	1.52
Bi2.5NIG(0.75)G	2.63	1.13	0.36	3.18	2.72	0.82	4.08	3.03	1.58
Bi2.5NIGG	2.63	1.18	0.37	3.19	3.18	0.86	4.16	3.28	1.49

Gaussian 4			Gaussian 5		
E <sub>0</sub> (eV)	Amp	Br (eV)	E <sub>0</sub> (eV)	Amp	Br (eV)
4.06	2.69	1.39	5.70	3.20	3.16
4.02	2.92	1.40	5.72	3.18	3.06
5.48	1.41	1.43	6.98	2.69	2.40
5.31	1.81	2.26	8.02	2.5	4.59
4.94	2.04	1.21	6.24	3.53	3.28
5.40	3.69	2.30	7.94	5.66	3.61
5.72	3.88	2.82	7.22	2.76	2.59
6.09	5.22	2.48	7.05	6.41	4.21

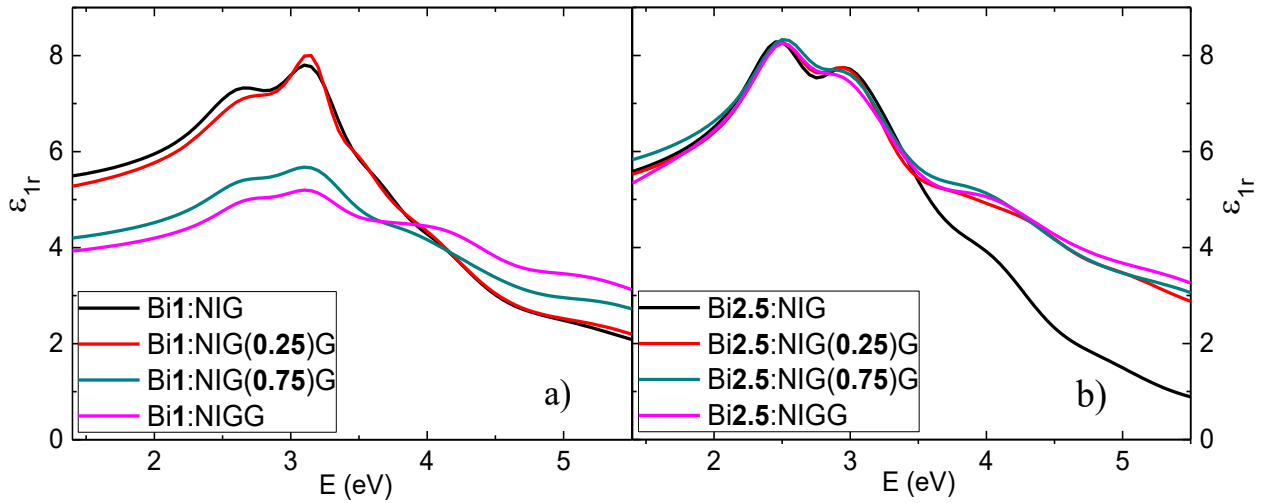
**Table 6.2.7: Fitted thicknesses and volume fractions used for model of Bi1:NiGxGs and Bi2.5:NiGxGs layers on GGG substrate in the spectral range from 1.5 to 6.5 eV. Here, T<sub>Bi:NiGG</sub> stands for film thickness; R<sub>rough</sub> represents film roughness with volume fractions f<sub>rough</sub>.**

	T <sub>Bi:NiGG</sub> (nm)	R <sub>rough</sub> (nm)	f <sub>rough</sub>
Bi1NiG	168.5	0	0.5
Bi1NiG(0.25)G	205	0	0.5
Bi1NiG(0.75)G	250	0	0.5
Bi1NiGG	265	0	0.5
Bi2.5NiG	150	10	0.5
Bi2.5NiG(0.25)G	152	5.2	0.5
Bi2.5NiG(0.75)G	155	4.6	0.5
Bi2.5NiGG	157	5	0.5

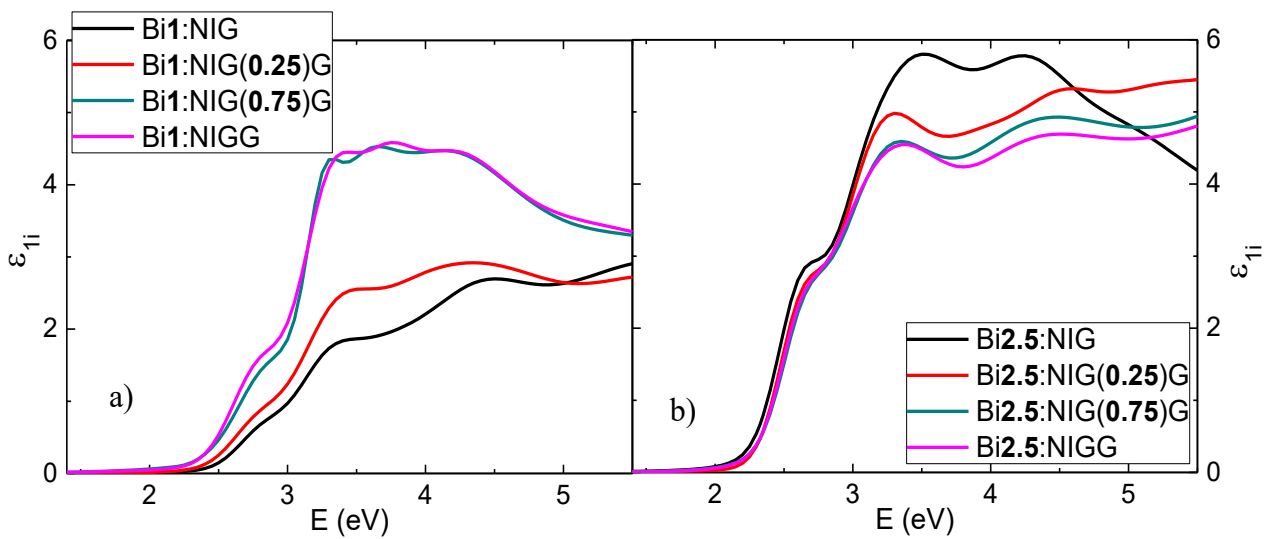
Figure 6.2.18(a) shows parameterized optical functions  $\epsilon_{1r}$  of Bi1:NiGxGs. Figure 6.2.18(b) shows parameterized optical functions  $\epsilon_{1r}$  of Bi2.5:NiGxGs. From these spectra, it is apparent that Ga substitution decreases amplitudes of  $\epsilon_{1r}$  for Bi1:NiGxGs below 4 eV and increasing them above. On the other hand, Ga substitution does not noticeably influence amplitudes of  $\epsilon_{1r}$  for Bi2.5:NiGxGs. The only exception is  $\epsilon_{1r}$  spectra of Bi2.5:NiG above 3.5 eV. We attribute this result to the fact that this composition contains no Ga.

Figure 6.2.19(a) shows parameterized optical functions  $\epsilon_{1i}$  of Bi1:NiGxGs. Figure 6.2.19(b) shows parameterized optical functions  $\epsilon_{1i}$  of Bi2.5:NiGxGs. In here, we can observe that Ga substitution increases  $\epsilon_{1i}$  amplitudes and therefore an absorption

of Bi1:NIGxGs in the measured spectral range. However it decreases  $\epsilon_{li}$  amplitudes of Bi2.5:NIGxGs. Moreover, the absorption of Bi2.5:NIGxGs is almost 30% stronger than the absorption of Bi1:NIGxGs. We attribute this result to the higher Bi concentration, since this effect of Bi was previously demonstrated in the part devoted to Bi<sub>x</sub>:YIGs.



**Figure 6.2.18: Real parts of the diagonal permittivity tensor elements  $\epsilon_{1r}$  for a) Bi1:NIGxGs and b) Bi2.5:NIGxGs.**

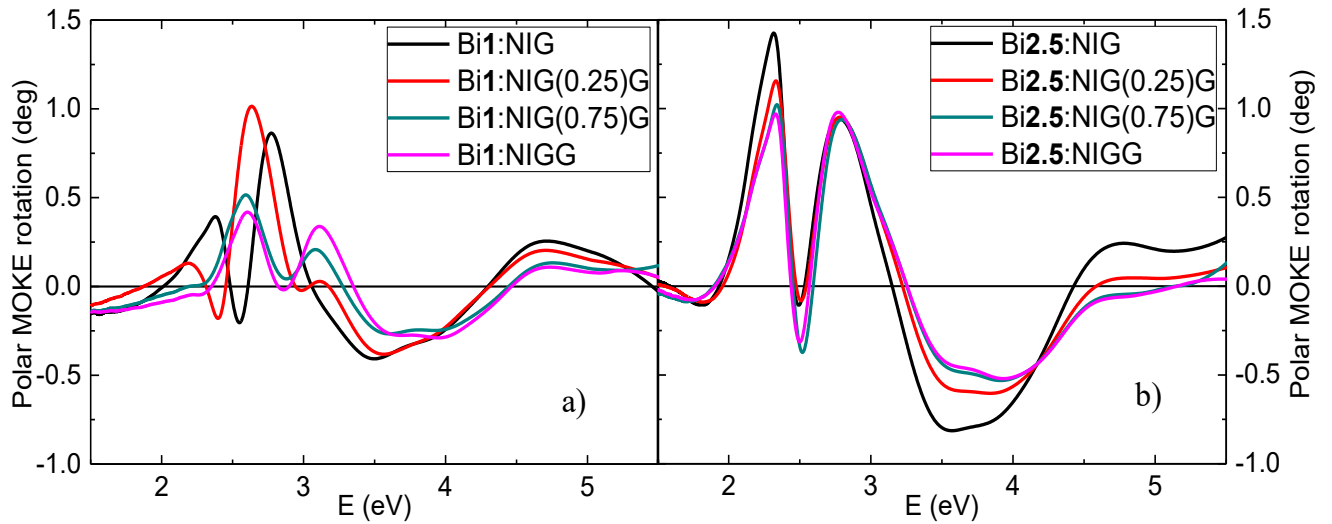


**Figure 6.2.19: Imaginary parts of the diagonal permittivity tensor elements  $\epsilon_{1i}$  for a) Bi1:NIGxGs and b) Bi2.5:NIGxGs.**

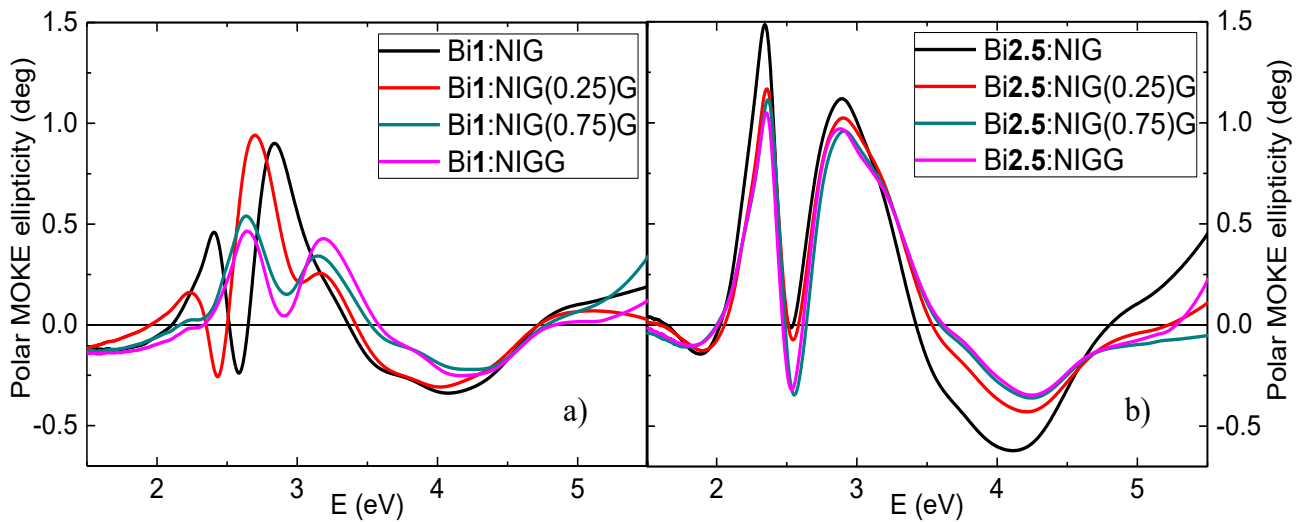
## b) MOKE and Faraday effect spectroscopy

We studied MO properties of Bi1:NiGxGs and Bi2.5:NiGxGs by MOKE and MO Faraday effect spectroscopy. We measured spectra of polar MOKE rotation and ellipticity at room temperature at nearly normal light incidence. We applied magnetic field 1.2 T, which was enough for samples saturation. Incident light was p polarized. We recorded data in the photon energy range from 1.4 to 5 eV. Similarly, we measured spectra of Faraday rotation and ellipticity at room temperature using magnetic field 670 mT (enough for samples saturation). We recorded experimental data in the photon energy range from 1.4 to 4 eV.

Figure 6.2.20(a) shows measured MOKE rotation spectra of Bi1:NiGxGs and Figure 6.2.20(b) of Bi2.5:NiGxGs. Figure 6.2.21(a) shows measured MOKE ellipticity spectra of Bi1:NiGxGs and Figure 6.2.21(b) of Bi2.5:NiGxGs. Firstly, we can observe strong MO interference in the form of strong oscillations in the spectral range below 3 eV for Bi1:NiGxGs and Bi2.5:NiGxGs in both, rotation and ellipticity. Since our samples had different thicknesses, we observed different interference patterns for each one of them (see Table 6.2.7). Secondly, MOKE rotation shows extremes around 3.5 and 4.5 eV for both sample sets. However, it is apparent that Bi2.5:NiGxGs shows higher MOKE amplitudes at extremes caused by the higher Bi content [8, 82]. Finally, MOKE ellipticity shows extreme around 4.1 eV for both sample sets which is characteristic for iron garnets MOKE spectra [8, 68, 82]. Important observation here is that Ga substitution decreases MOKE rotation amplitudes at extremes. As expected, spectra demonstrated that Bi substitution increases, and Ga substitution decreases amplitudes of MOKE ellipticity.



**Figure 6.2.20: Polar MOKE rotation spectra for a) Bi1:NIGxGs and b) Bi2.5:NIGxGs on GGG substrates.**



**Figure 6.2.21: Polar MOKE ellipticity spectra for a) Bi1:NIGxGs and b) Bi2.5:NIGxGs on GGG substrates.**

Figure 6.2.22(a) shows measured MO Faraday rotation spectra for Bi1:NIGxGs and Figure 6.2.22(b) for Bi2.5:NIGxGs. Figure 6.2.23(a) shows measured MO Faraday ellipticity spectra for Bi1:NIGxGs and Figure 6.2.23(b) for Bi2.5:NIGxGs. We corrected experimental data for the rotation from the substrate. We observed Faraday rotation extremes near 2.5 and 3 eV, and Faraday ellipticity extremes near 2.3 and 3.3 eV for both sample sets. Spectra clearly demonstrated that Bi2.5:NIGxGs shows

higher Faraday rotation and ellipticity amplitudes at extremes, caused by the higher Bi content [8, 68, 82]. One important thing to notice is that Ga substitution decreases MO Faraday effect amplitudes at extremes. This effect of Ga will be discussed later in the part devoted to the microscopic analysis of studied materials.

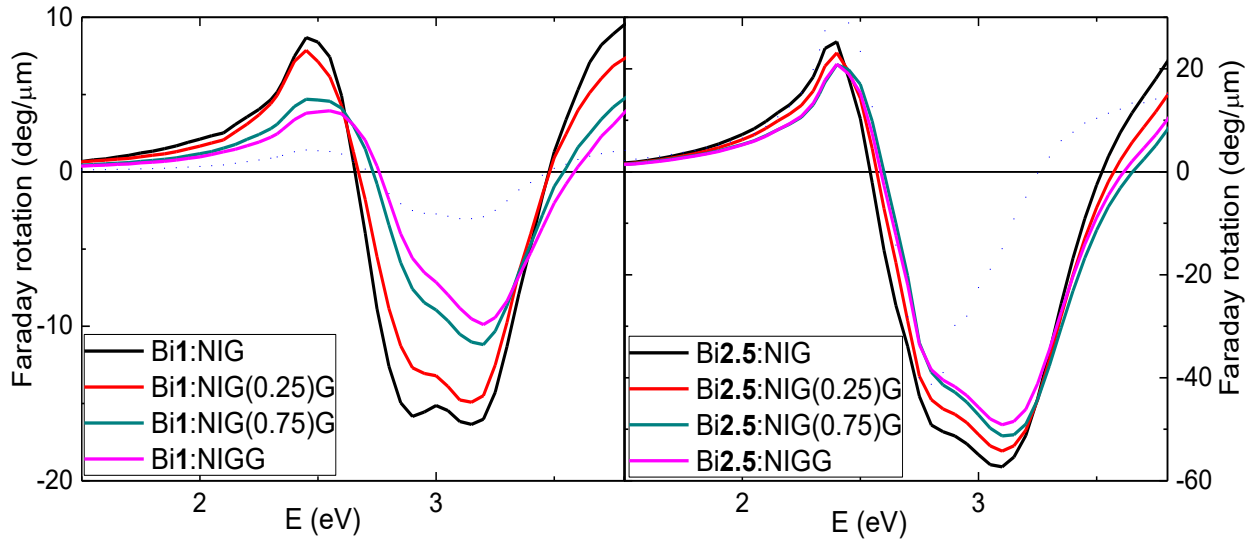


Figure 6.2.22: MO Faraday rotation spectra for a) Bi1:NiG<sub>x</sub>Gs and b) Bi2.5:NiG<sub>x</sub>Gs on GGG substrates.

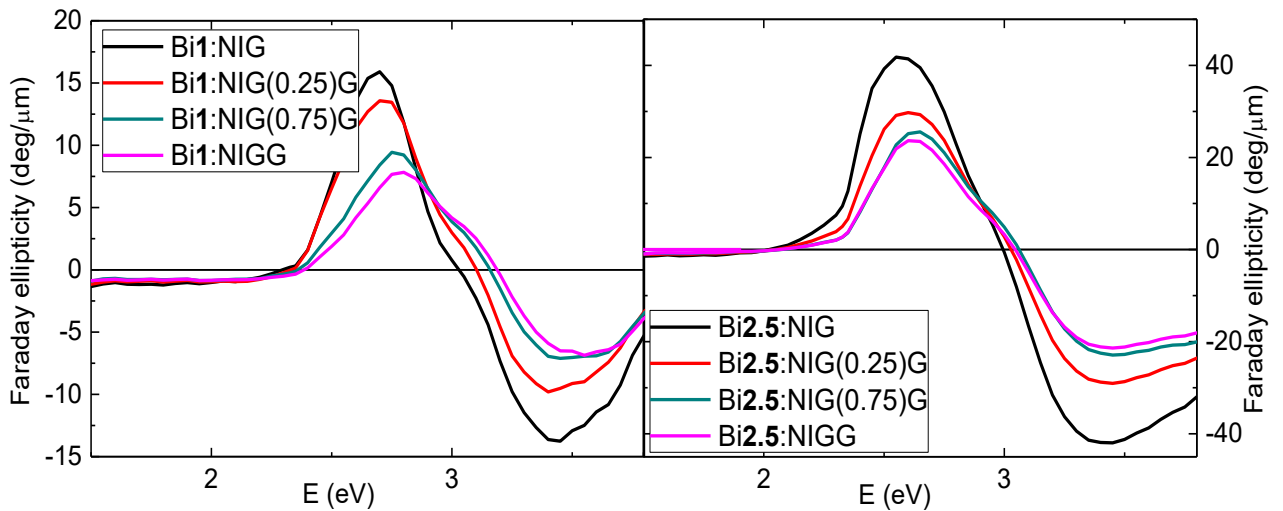


Figure 6.2.23: MO Faraday ellipticity spectra for a) Bi1:NiG<sub>x</sub>Gs and b) Bi2.5:NiG<sub>x</sub>Gs on GGG substrates.

We calculated off-diagonal elements of the permittivity tensor  $\epsilon_{2r}$  and  $\epsilon_{2i}$  from MOKE and Faraday effect spectra. We performed calculations using Yeh's 4x4

matrix formalism and diagonal elements of the permittivity tensor determined by SE. As already mentioned, there was strong MO interference below 3 eV in MOKE spectra. Therefore, we used MO Faraday effect spectra in the spectral range from 1.5 to 3 eV and MOKE spectra in the spectral range from 3 to 5 eV. Figure 6.2.24(a) shows real parts of off-diagonal permittivity tensor elements  $\epsilon_{2r}$  of Bi1:NIGxGs and Figure 6.2.24(b) of Bi2.5:NIGxGs. Figure 6.2.25(a) shows imaginary parts of off-diagonal permittivity tensor elements  $\epsilon_{2i}$  of Bi1:NIGxGs and Figure 6.2.25(b) of Bi2.5:NIGxGs. As expected from previous results, Bi substitution increases  $\epsilon_{2r}$  and  $\epsilon_{2i}$  amplitudes at extremes for both, Bi1:NIGxGs and Bi2.5:NIGxGs. However, Ga substitution acts in an opposite manner and it decreases  $\epsilon_{2r}$  and  $\epsilon_{2i}$  amplitudes.

To explain the effect of Ga properly, one has to look at the results from microscopic theory. For this purpose, we parameterized off-diagonal elements spectra of Bi1:NIGxGs and Bi2.5:NIGxGs permittivity tensors,  $\epsilon_{2r}$  and  $\epsilon_{2i}$ . We listed some of used transitions parameters in table 6.2.4. We used four Para transition terms at 2.4-2.5, 3-3.2, 3.6-4.1 and 5.7-6.6 eV to represent crystal field transitions. Furthermore, we used four Dia transition terms at 0.5, 2.58-2.8, 3.3-3.58 and 4.05-4.4 eV. We used Dia transition at 0.5 eV and Para transition at 5.7-6.6 eV only to model combined effect of transitions outside of measured spectral range and we do not assign them any physical meaning in here. First thing to notice is that the main contribution comes, similarly to Bi<sub>x</sub>:YIGs, from Dia transitions at 2.58-2.8 eV and 3.3-3.58 eV. These are associated with transitions  $t_2(\text{Fe}^{3+}) \rightarrow t_2g(\text{Fe}^{2+})$  and  $eg(\text{Fe}^{3+}) \rightarrow e(\text{Fe}^{2+})$  respectively. As mentioned before, these transitions are mainly responsible for the remarkable increase of the Faraday rotation in the visible and near infrared region. From the data in the table 6.2.4 one can see that Ga substitution is decreasing these transitions for both materials. This is in accordance with the assumption, that Ga is mostly substituted for Fe<sup>3+</sup> tetrahedral, which is crucial for both transitions. Maxima at 2.4 eV are mainly created by the overlap of secondary negative peaks of these two Dia transitions. One can also notice that Ga substitution lowers energy of much smaller Dia transition at 4.65 eV which most likely exist due to charge transfers from oxygen to octahedral Fe. This is in accordance with the assumption that Ga is in a smaller percentage also substituted per Fe<sup>3+</sup> octahedral.



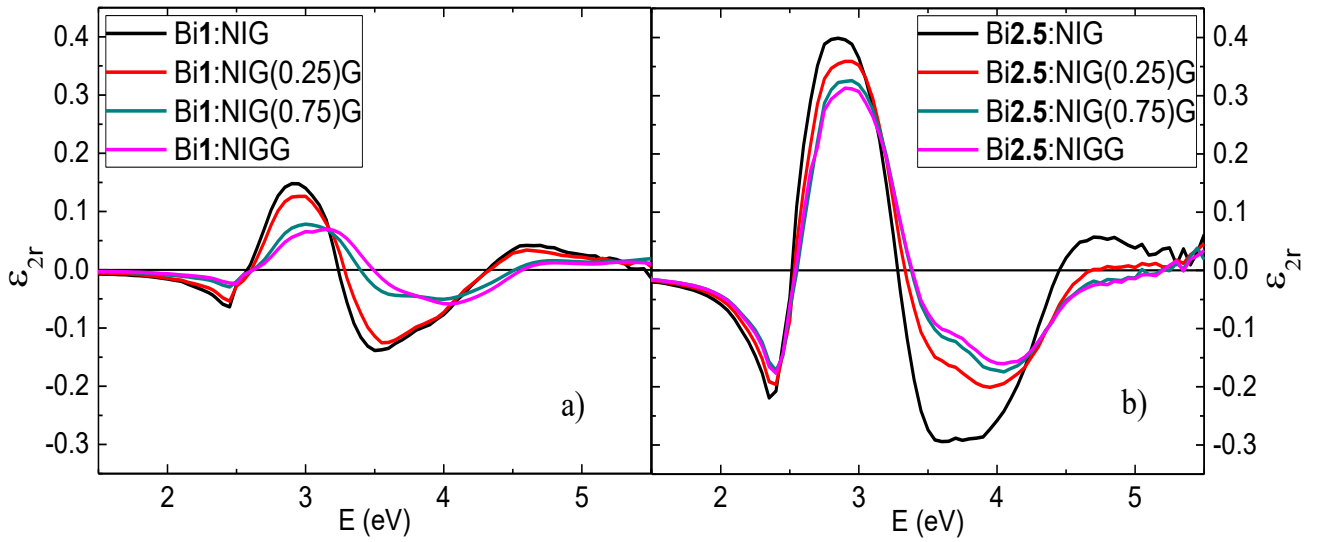


Figure 6.2.24: Real parts of the off-diagonal permittivity tensor elements  $\epsilon_{2r}$  for a) Bi<sub>1</sub>:NiG<sub>x</sub>Gs and b) Bi<sub>2.5</sub>:NiG<sub>x</sub>Gs.

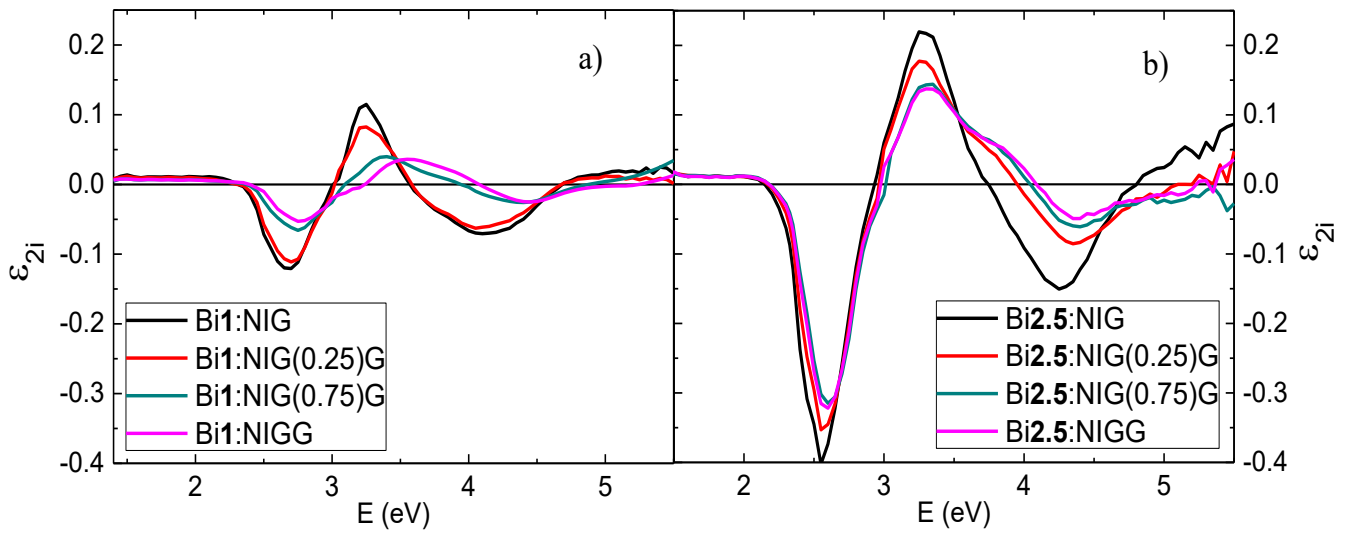


Figure 6.2.25: Real parts of the off-diagonal permittivity tensor elements  $\epsilon_{2i}$  for a) Bi<sub>1</sub>:NiG<sub>x</sub>Gs and b) Bi<sub>2.5</sub>:NiG<sub>x</sub>Gs.

**Table 6.2.8: Fitted parameters of Para and Dia transitions used to parameterize off-diagonal elements of the permittivity tensors of Bi1:NiGxGs and Bi2.5:NiGxGs layers prepared on GGG (111) substrates in the spectral range from 1.5 to 5.5 eV. Here,  $E_0$  stands for central energy of the function; Amp represents amplitude of the function and  $\Gamma_0$  is half-width in a half-height of the transition.**

sign	Dia tr. 1			Dia tr. 2			Dia tr. 3			Dia tr. 4		
	1			-1			1			-1		
	$E_0$ (eV)	Amp	$\Gamma_0$ (eV)	$E_0$ (eV)	Amp	$\Gamma_0$ (eV)	$E_0$ (eV)	Amp	$\Gamma_0$ (eV)	$E_0$ (eV)	Amp	$\Gamma_0$ (eV)
Bi1NiG	0.5	0.04	0.4	2.67	0.14	0.2	3.24	0.2	0.2	4.1	0.1	0.6
Bi1NiG(0.25)G	0.5	0.03	0.4	2.68	0.14	0.2	3.25	0.16	0.25	4.05	0.09	0.5
Bi1NiG(0.75)G	0.5	0.03	0.4	2.75	0.08	0.2	3.35	0.07	0.3	4.39	0.04	0.4
Bi1NiGG	0.5	0.03	0.4	2.8	0.065	0.3	3.58	0.065	0.4	4.4	0.05	0.4
Bi2.5NiG	0.5	0.03	0.4	2.58	0.55	0.5	3.3	0.38	0.5	4.25	0.28	0.5
Bi2.5NiG(0.25)G	0.5	0.03	0.4	2.6	0.45	0.17	3.3	0.28	0.5	4.4	0.17	0.55
Bi2.5NiG(0.75)G	0.5	0.03	0.4	2.6	0.43	0.17	3.3	0.24	0.5	4.4	0.12	0.55
Bi2.5NiGG	0.5	0.03	0.4	2.6	0.42	0.17	3.3	0.23	0.5	4.4	0.12	0.55

Para tr. 1			Para tr. 2			Para tr. 3			Para tr. 4		
-1			1			-1			1		
$E_0$ (eV)	Amp	$\Gamma_0$ (eV)	$E_0$ (eV)	Amp	$\Gamma_0$ (eV)	$E_0$ (eV)	Amp	$\Gamma_0$ (eV)	$E_0$ (eV)	Amp	$\Gamma_0$ (eV)
2.5	0.005	0.3	3	0.025	0.3	3.6	0.03	0.6	5.8	0.03	0.6
2.5	0.005	0.3	3	0.015	0.3	3.6	0.03	0.6	5.8	0.03	0.7
2.5	0.004	0.2	3.1	0.01	0.3	3.8	0.02	0.6	6	0.034	0.78
2.5	0.004	0.3	3.2	0.01	0.2	4	0.02	0.6	6.6	0.035	0.8
2.4	0.07	0.2	3.1	0.009	0.3	3.9	0.06	0.6	5.7	0.06	0.5
2.4	0.05	0.2	3.1	0.009	0.3	4.1	0.07	0.6			
2.4	0.05	0.2	3.2	0.008	0.2	4.1	0.08	0.6			
2.4	0.03	0.2	3.2	0.008	0.3	4.1	0.06	0.6			

### 6.3. $Ce_{(0.95-x)}Hf_xCo_{0.05}O_{(2-\delta)}$

In recent years, magnetically doped  $CeO_2$  attracted a lot of attention since it is a promising magnetic semiconductor and highly applicable material in the field of integrated photonics. This ranges from MO applications such as integrated MO isolators or magneto-plasmonic sensors to magneto-photonic crystals [83-86]. The main advantage of this material is its high Curie temperature and more importantly a

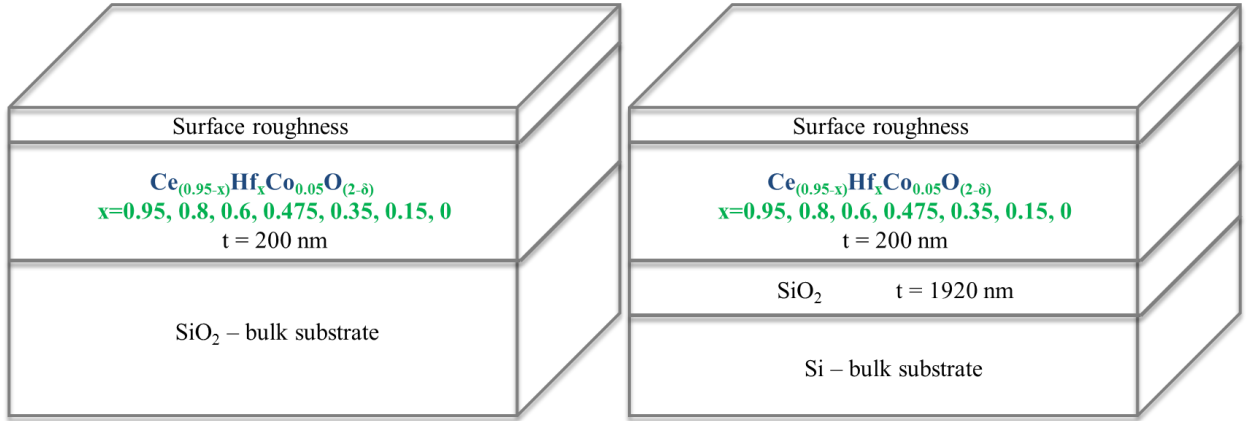
great Si compatibility [87]. Moreover, it is possible to tune CeO<sub>2</sub> magnetic properties by doping of the non-magnetic lattice by magnetic ions [83]. In this work we used Hf and Co doping. Successful adoption of this material in MO devices requires complete understanding of the nature and origin of CeO<sub>2</sub> magnetic properties. Even though is the room temperature ferromagnetism in this material explained by an oxygen vacancy mechanism [88, 89], the detail optical and MO analysis is still needed.

In this work, we focused mainly on the determination of full dielectric permittivity tensors of Ce<sub>(0.95-x)</sub>Hf<sub>x</sub>Co<sub>0.05</sub>O<sub>(2-δ)</sub> (CeHfCoO) thin films. For this purpose, we used optical and MO spectral measurements at energies from 1.5 to 5.5 eV. We used SE supported by transmission intensity measurements to study CeHfCoO optical properties and to derive the diagonal permittivity tensor elements spectra. We used spectroscopic Faraday effect rotation and ellipticity measurements to study CeHfCoO MO properties and to derive the spectral dependence of off-diagonal permittivity tensor elements. Finally, we parameterized obtained results in terms of microscopic theory.

We studied polycrystalline Ce<sub>(0.95-x)</sub>Hf<sub>x</sub>Co<sub>0.05</sub>O<sub>(2-δ)</sub> thin films (x = 0, 0.15, 0.35, 0.475, 0.6, 0.8, 0.95) prepared by pulsed laser deposition method on 2 types of substrates: amorphous quartz and Si/SiO<sub>2</sub>. The deposition was carried out in vacuum (at base pressure 1.0x10<sup>-6</sup> Torr) with substrate temperature of 700<sup>0</sup>C. We listed CeHfCoO thin films compositions and nominal thicknesses (determined by profilometer) in table 6.3.1. Figure 6.3.1 shows theoretical model structure of CeHfCoO samples used for SE and Faraday effect analysis.

**Table 6.3.1: Composition and nominal thicknesses of examined CeHfCoO thin films prepared on amorphous quartz and Si/SiO<sub>2</sub> substrates.**

Film composition	Indication in Figures and text	Nominal thickness (nm)
Ce <sub>0.95</sub> Co <sub>0.05</sub> O <sub>(2-δ)</sub>	Ce0.95CoO	310
Ce <sub>0.8</sub> Hf <sub>0.15</sub> Co <sub>0.05</sub> O <sub>(2-δ)</sub>	CeHf0.15CoO	310
Ce <sub>0.6</sub> Hf <sub>0.35</sub> Co <sub>0.05</sub> O <sub>(2-δ)</sub>	CeHf0.35CoO	310
Ce <sub>0.475</sub> Hf <sub>0.475</sub> Co <sub>0.05</sub> O <sub>(2-δ)</sub>	CeHf0.475CoO	340
Ce <sub>0.35</sub> Hf <sub>0.60</sub> Co <sub>0.05</sub> O <sub>(2-δ)</sub>	CeHf0.60CoO	310
Ce <sub>0.15</sub> Hf <sub>0.80</sub> Co <sub>0.05</sub> O <sub>(2-δ)</sub>	CeHf0.80CoO	300
Hf <sub>0.95</sub> Co <sub>0.05</sub> O <sub>(2-δ)</sub>	Hf0.95CoO	150



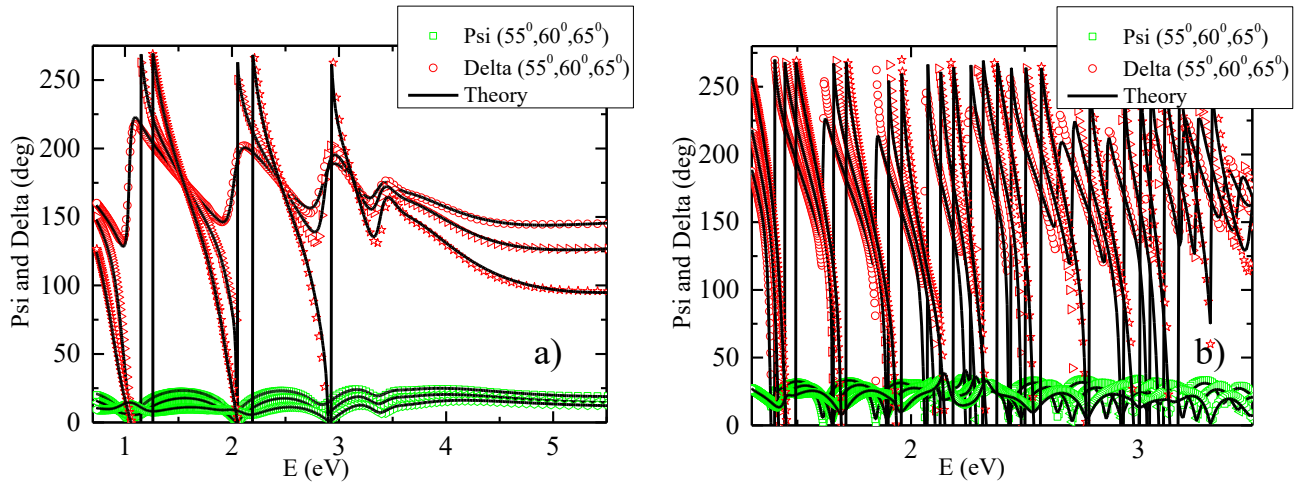
**Figure 6.3.1: Model structure of CeHfCoO samples used for SE and Faraday analysis calculations.**

### a) Spectroscopic Ellipsometry

We performed SE measurements on a Mueller matrix ellipsometer Woollam RC2. We measured spectral dependence of ellipsometry parameters  $\psi$  and  $\Delta$  in reflection and at incident angles  $55^\circ$ ,  $60^\circ$  and  $65^\circ$ . We used the same equipment to measure the transmission spectra of CeHfCoO thin films prepared on transparent amorphous quartz substrates and we used incidence angle  $0^\circ$ . We performed measurements in the spectral range from 1.5 to 6.5 eV.

We analyzed SE experimental data using a CompleteEase software provided by Woollam Co.. We used MSA mode to obtain optical functions spectra (diagonal elements of the permittivity tensor  $\epsilon_{1r}$  and  $\epsilon_{1i}$ ) of CeHfCoO thin films. In MSA mode, we combined SE and transmission measurements for each CeHfCoO film composition on both substrates. This means that for each composition we took experimental data from CeHfCoO sample prepared on quartz substrate ( $\psi$ ,  $\Delta$  and transmission experimental spectra) and combined them with experimental data from CeHfCoO sample prepared on Si/SiO<sub>2</sub> substrate ( $\psi$  and  $\Delta$  experimental spectra). In MSA, we treated CeHfCoO, Si and SiO<sub>2</sub> optical functions as parameters common for both samples. We treated all thicknesses (including roughness thickness) as parameters allowed to vary for each sample independently. This approach provided us with the more accurate results since it ensured that obtained constants describe all types of experimental data well. Figure 6.3.2 shows that used theoretical model

describes both SE experimental spectra well for both substrates. As a final SE step, we parameterized derived diagonal elements of the permittivity tensor  $\epsilon_{1r}$  and  $\epsilon_{1i}$  using a combination of Lorentz, Gaussian, and Tauc-Lorentz oscillators. We did this to obtain Kramers-Kronig consistent results. We listed some parameters of used oscillator functions in Table 6.3.2 and the fitted thicknesses in Table 6.3.3. We subsequently used all fitted thicknesses (including roughness) obtained from SE analysis in Faraday effect analysis.



**Figure 6.3.2: Measured SE variable angle Psi and Delta spectra of a) CeHf<sub>0.15</sub>CoO prepared on Si/SiO<sub>2</sub> substrate and b) CeHf<sub>0.15</sub>CoO prepared on quartz substrate compared to the theoretical model.**

**Table 6.3.2: Fitted parameters of oscillator functions used to parameterize optical properties of CeHfCoO thin films prepared on amorphous quartz and Si/SiO<sub>2</sub> substrates in the spectral range from 1.7 to 5 eV. Here, E<sub>0</sub> stands for central energy of the function; Amp represents amplitude of the function and Br its broadening.**

	Lorentz 1			Lorentz 2			Lorentz 3		
	E <sub>0</sub> (eV)	Amp	Br (eV)	E <sub>0</sub> (eV)	Amp	Br (eV)	E <sub>0</sub> (eV)	Amp	Br (eV)
Ce <sub>0.95</sub> CoO	5.37	1.94	1.65	6.34	3.36	0.9	15	2.9	9.48
CeHf <sub>0.15</sub> CoO	5.19	1.45	0.93	6.25	1.7	1.87	14	8.65	5.03
CeHf <sub>0.35</sub> CoO	4.83	1.67	0.9	5.83	1.71	2.09	14.96	9.45	4.85
CeHf <sub>0.475</sub> CoO	4.75	1.92	0.76	5.49	1.32	1.31	12.52	10	4.65
CeHf <sub>0.60</sub> CoO	4.61	2.35	0.8	5.44	1.73	1.64	10.67	11.06	3.03
CeHf <sub>0.80</sub> CoO	4.62	2.99	0.88	5.92	2.55	1.99	9.79	15.89	1.69
Hf <sub>0.95</sub> CoO	-	-	-	-	-	-	15	32.11	1.38

Gaussian			Tauc Lorentz		
E <sub>0</sub> (eV)	Amp	Br (eV)	E <sub>0</sub> (eV)	Amp	Br (eV)
5.58	0.94	1.19	15	14.09	0.0001
4.48	1.78	0.68	-	-	-
4.25	2.25	0.59	-	-	-
4.15	3.42	0.67	-	-	-
4.02	4.17	0.65	-	-	-
3.98	6.61	0.56	-	-	-
7.34	4.52	2.2	-	-	-

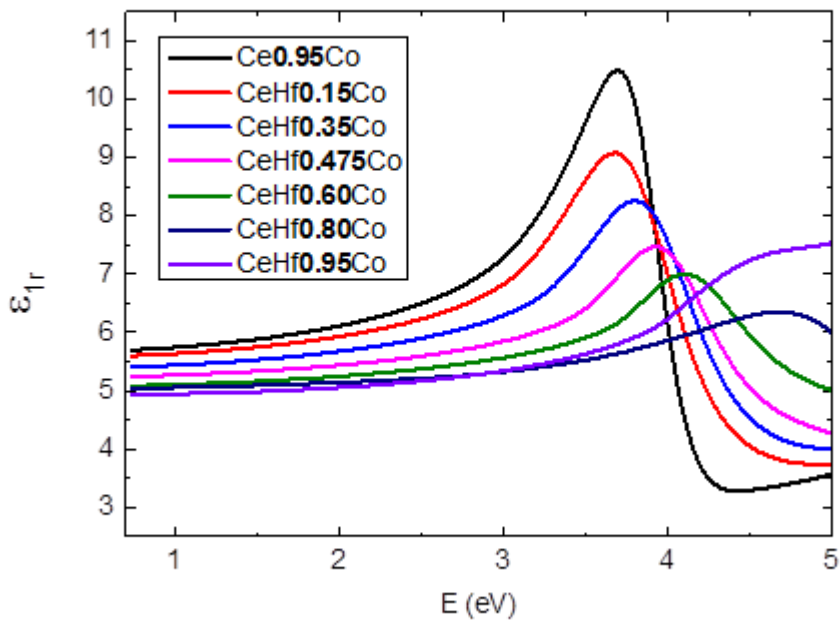
**Table 6.3.3: Fitted thicknesses and volume fractions used for model of CeHfCoO layers on quartz and Si/ SiO<sub>2</sub> substrate in the spectral range from 1.7 to 5 eV. Here, T/quartz and R/quartz stand for thickness and roughness of CeHfCoO films prepared on quartz substrate respectively; T/Si and R/Si stand for thickness and roughness of CeHfCoO films prepared on Si substrate.**

	T /quartz (nm)	R /quartz (nm)	T/Si (nm)	R/Si (nm)
Ce0.95CoO	180.9	2	217.6	1
CeHf0.15CoO	234.7	1	237.7	0.5
CeHf0.35CoO	437.2	0.5	410.9	0.6
CeHf0.475CoO	438.9	0.5	365.7	0.5
CeHf0.60CoO	359.2	0.4	316	0.5
CeHf0.80CoO	269.3	2.5	242	0.5
Hf0.95CoO	204.7	2.3	122.6	3.7

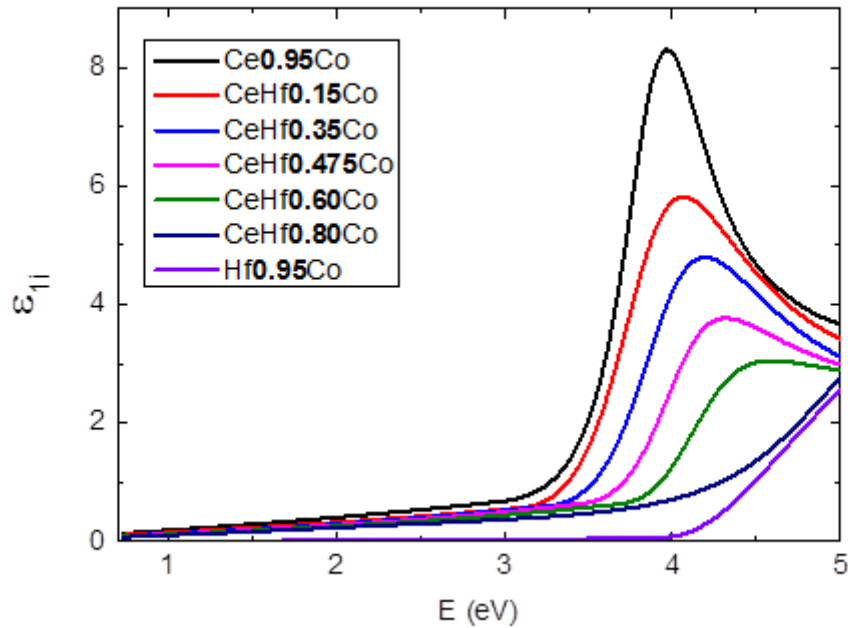
Figure 6.3.3 shows real parts of diagonal permittivity tensor elements  $\epsilon_{1r}$  of CeHfCoO films. The spectral dependence of  $\epsilon_{1r}$  is similar in shape to results obtained on pure or Co doped CeO<sub>2</sub> films [90-92]. We can observe that all the spectra are characteristic by one global maximum shifting from 3.6 eV to higher energies when Hf content increasing. Moreover, it is apparent that Hf content decreases  $\epsilon_{1r}$  amplitudes in the whole measured spectral range. This is caused by smaller absorption. One extra thing to observe are slightly higher amplitudes for Hf0.95Co material then expected (above 3 eV) from the trend that shows other compositions when increasing Hf content. We assume that this is probably caused by missing Co in the structure.

Figure 6.3.4 shows imaginary parts of the diagonal permittivity tensor elements  $\epsilon_{1i}$  of CeHfCoO thin films. In here we can observe optical bandgap energies to be shifted

from 3.21 to 4.1 eV when Hf content increases. These energies are related to O2p  $\rightarrow$  Ce4f electronic transitions [92-94]. Moreover, one can see that Hf doping decreases  $\epsilon_{li}$  amplitudes in the whole measured spectral range. From this result we assume that when is Hf replacing Ce in the material; it acts against optical vacancies from isolated Ce4f states localized within the optical bandgap. These vacancies serves as recombination centers for optically excited electrons from the valence band to the 4f band of the oxide and are responsible for enhanced optical absorption [92, 93, 95]. One more thing to support this theory is increased absorption tail below 3.2 eV for our samples. This absorption tail was previously explained by the effect of mid-gap defects (mid-gap oxygen vacancies, Co states) [92] and it can be clearly seen that this tail disappearing when Hf content increases.



**Figure 6.3.3: Real parts of the diagonal permittivity tensor elements  $\epsilon_{1r}$  for CeHfCoO thin films.**



**Figure 6.3.4: Imaginary parts of the diagonal permittivity tensor elements  $\epsilon_{11}$  for CeHfCoO thin films.**

### **b) Faraday effect spectroscopy**

We studied MO properties of examined samples by MO Faraday effect spectroscopy. We performed this type of measurement only on samples with quartz substrate since these samples are transparent in the whole measured spectral range. Therefore, there was no need for additional MOKE measurement. We acquired all the spectra at room temperature and normal light incidence. We applied magnetic field 670 mT, which was enough for samples saturation. Incident light was p polarized. We recorded data in the photon energy range from 0.7 to 4 eV.

Figure 6.3.5 shows measured MO Faraday rotation spectra of CeHfCoO films. Figure 6.3.6 shows measured MO Faraday ellipticity spectra of CeHfCoO films. We corrected experimental data for the rotation from the substrate. We observed Faraday rotation extreme shifting from 2.9 eV to higher energies and decreased rotation amplitudes when Hf content increased. On the other hand, ellipticity amplitudes increased their values when Hf content increased. Spectra of the fully Hf substituted



sample Hf0.95CoO has different rotation and ellipticity amplitudes than expected from the trend seen on other samples when Hf concentration increases. We attribute this to the missing Ce in the structure.

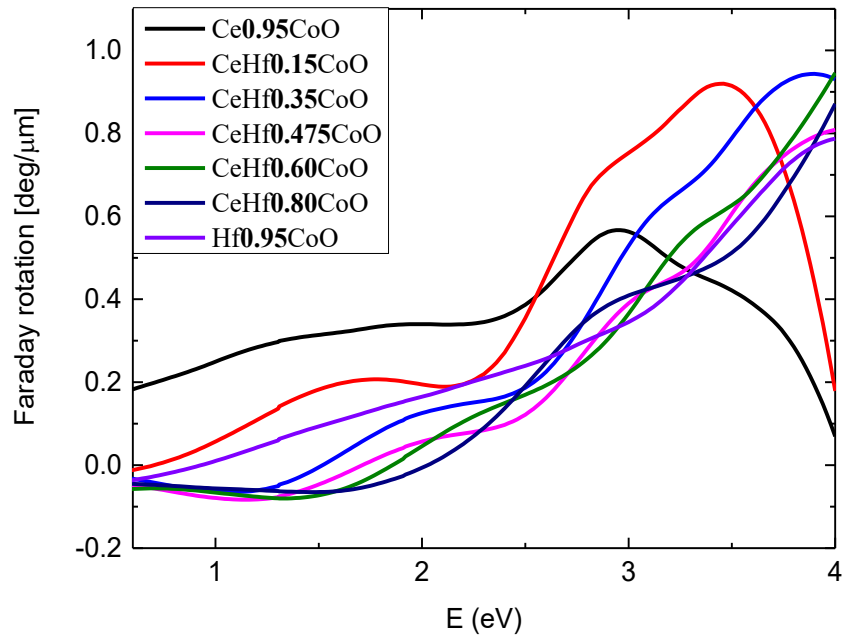


Figure 6.3.5: MO Faraday rotation spectra of CeHfCoO films prepared on quartz substrates.

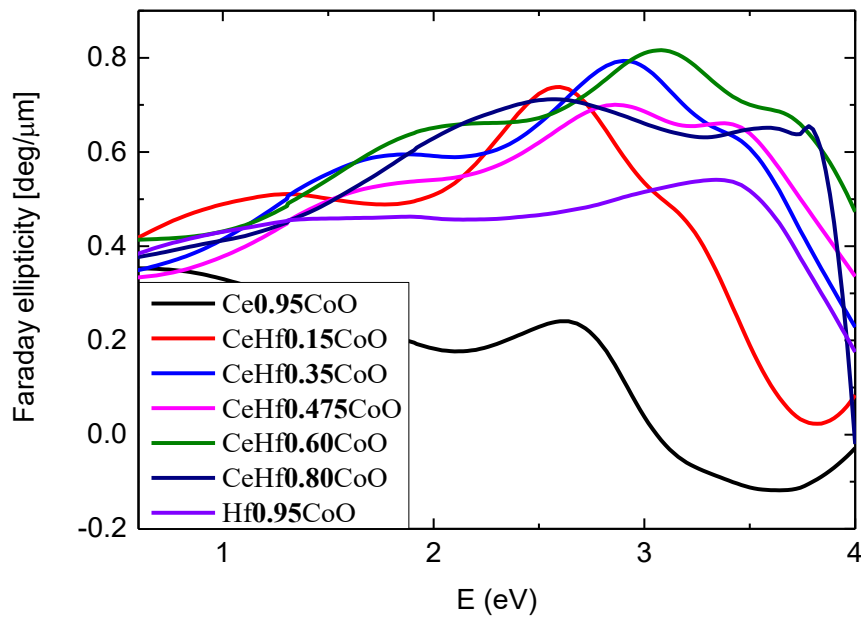


Figure 6.3.6: MO Faraday ellipticity spectra of CeHfCoO films prepared on quartz substrates.

We calculated the off-diagonal elements of the permittivity tensor  $\epsilon_{2r}$  and  $\epsilon_{2i}$  from Faraday effect spectra using Yeh's 4x4 matrix formalism and diagonal elements of the permittivity tensor determined by SE. Figure 6.3.7 shows the real parts of the off-diagonal permittivity tensor elements  $\epsilon_{2r}$ . Figure 6.3.8 shows the imaginary parts of the off-diagonal permittivity tensor elements  $\epsilon_{2i}$ . Spectra clearly demonstrated that Hf substitution decreases both,  $\epsilon_{2i}$  and  $\epsilon_{2r}$  amplitudes and shifting their maxima to higher energies in the measure spectral range. This can be explained by the fact that perpendicular magnetic anisotropy and room temperature ferromagnetism in this material is attributed to the magnetoelastic effects. These effects originate from distortions which are caused by in-plane compressive strain and vary mainly with Ce-Co content [92, 96]. Hf substitution not only influences this content, but it also reduces oxygen vacancies which also play an important role in CeHfCoO magnetism.

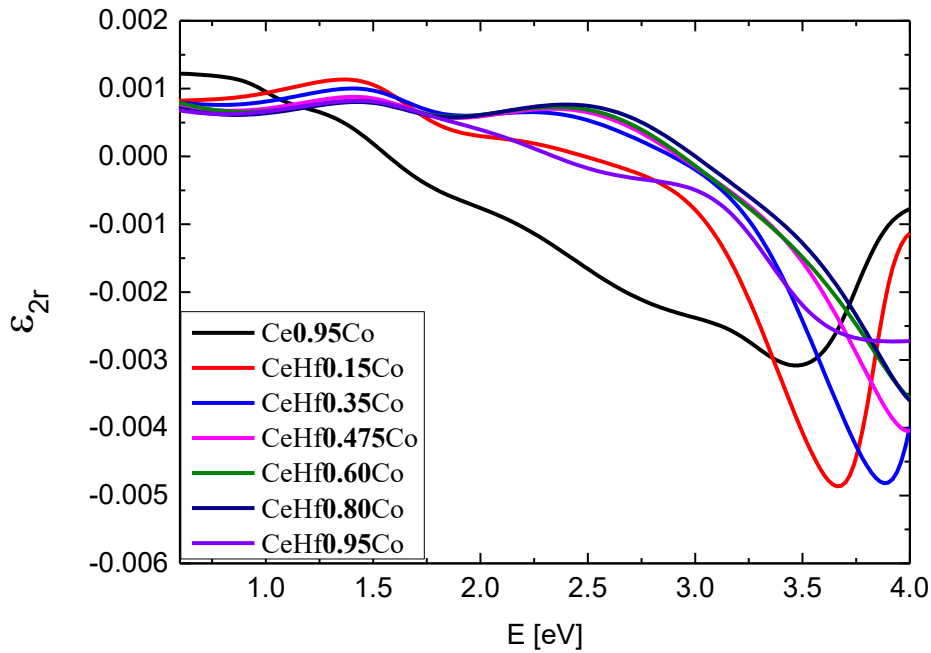
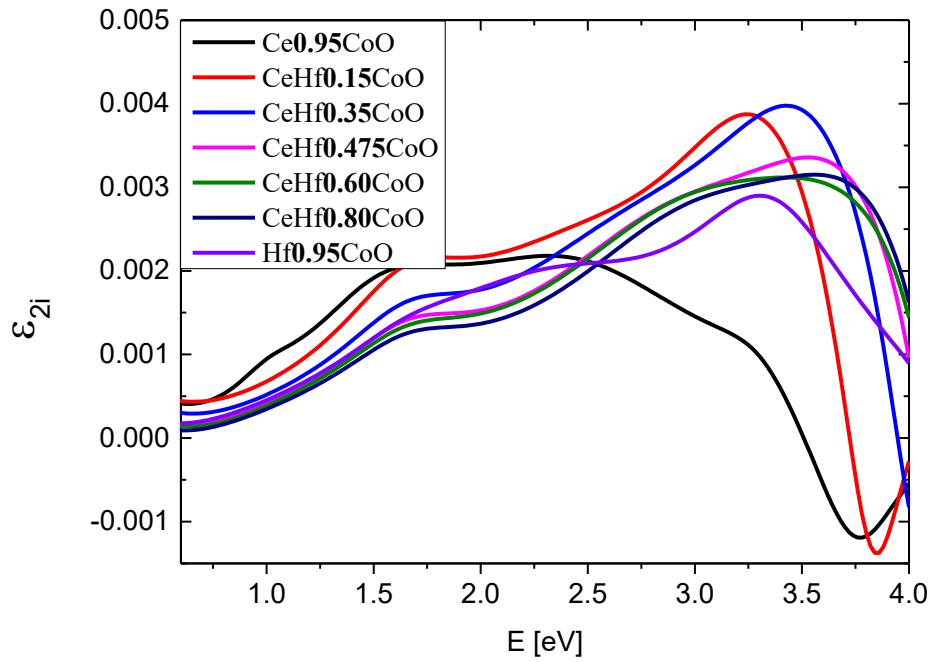


Figure 6.3.7: Real parts of the off-diagonal permittivity tensor elements  $\epsilon_{2r}$  of CeHfCoO films



**Figure 6.3.8: Imaginary parts of the off-diagonal permittivity tensor elements  $\epsilon_{2i}$  of CeHfCoO films.**

To relate calculated spectra to the microscopic theory, we parameterize them by the sum of Para and Dia oscillator terms. We listed some of used parameters in table 6.3.4. From the result, one can clearly see that the main MO contribution comes from Dia transitions 1 (1.5-1.65 eV), 2 (2.42-2.95 eV) and 4 (3.75-4.3 eV). These therefore correspond to excited state split by the combined effect of exchange field and spin-orbit coupling. Moreover, they are all decreasing their values with increased Hf content. Transition 1 refers to localized 4f states in the band gap while transition 4 to the oxygen electronic transitions [92].

**Table 6.3.4: Fitted parameters of Para and Dia transitions used to parameterize off-diagonal elements of the permittivity tensors of HfCoCoO films in the spectral range from 1.5 to 5.5 eV. Here,  $E_0$  stands for central energy of the function; Amp represents amplitude of the function and  $\Gamma_0$  is half-width in a half-height of the transition.**

sign	Dia tr 1			Dia tr 2			Dia tr 3		
	$E_0$ (eV)	Amp	$\Gamma_0$ (eV)	$E_0$ (eV)	Amp	$\Gamma_0$ (eV)	$E_0$ (eV)	Amp	Br (eV)
Ce0.95Co	1.53	0.0014	0.5	2.42	0.0022	0.9	3.3	0.0005	0.3
CeHf0.15Co	1.65	0.0011	0.4	2.4	0.0014	0.8	3.35	0.0038	0.6
CeHf0.35Co	1.65	0.0008	0.4	2.65	0.0015	0.8	3.55	0.0038	0.7
CeHf0.475Co	1.65	0.0007	0.4	2.85	0.0018	0.8	3.75	0.003	0.7
CeHf0.60Co	1.65	0.0006	0.4	2.9	0.0017	0.8	3.82	0.0027	0.9
CeHf0.80Co	1.65	0.0006	0.4	2.95	0.0019	0.8	3.85	0.0027	0.7
Hf0.95Co	1.65	0.0004	0.4	2.3	0.0017	0.85	3.35	0.0023	0.5

Dia tr 4			Dia tr 5			Para tr 1		
$E_0$ (eV)	Amp	$\Gamma_0$ (eV)	$E_0$ (eV)	Amp	$\Gamma_0$ (eV)	$E_0$ (eV)	Amp	$\Gamma_0$ (eV)
3.75	0.002	0.3	1	0.0003	0.2	0.35	0.0008	0.9
3.83	0.004	0.2	-	-	-	0.4	0.0004	0.7
4.05	0.004	0.2	-	-	-	0.35	0.0005	0.7
4.15	0.0035	0.2	-	-	-	0.3	0.0006	0.7
4.2	0.0033	0.2	-	-	-	0.3	0.0007	0.7
4.2	0.0032	0.2	-	-	-	0.27	0.0007	0.7
4.3	0.001	0.2	-	-	-	0.27	0.0006	0.7

## 7. CONCLUSIONS

In this part of the thesis, we would like to conclude our findings for all investigated materials.

To conclude our results on  $\text{Gd}_x\text{Fe}_{(100-x)}$ , it is important to note that perpendicular magnetic anisotropy is substantial for its application potential (MO-SLM, MO disk storage). This type of anisotropy is characteristic for concentrations close to  $x \approx 25$ , compensation concentration. We derived permittivity tensors for  $\text{Gd}_x\text{Fe}_{(100-x)}$  compositions close to this concentration. Our investigation showed that the behavior in the spectral range from 1.5 to 3eV, where  $\epsilon_{1r}$  decreases its value for higher energies, differs from typical Drude-like behavior (describing intra-band transitions) of metallic compounds. On the other hand, it is like the behavior of some transition metals. This is caused by the Lorentz contribution centered near 1.9 eV. This contribution most likely originates from the inter-band transition, which involves Fe 3d and Gd 5d states. The Fe 3d state is located around 1.5 eV below Fermi energy. Contrarywise, Gd 5d states are located approx. 0.5 eV above the Fermi energy. The second Lorentz oscillator near 2.5 eV does not significantly modify the spectral dependence of Drude behavior. The reason is its small amplitude. This points on the origin of Gd d-d electron transition, since this transition should be forbidden with small oscillator strength. Finally, we discuss the Gd substitution effect. The amplitude of the first Lorentz oscillator near 1.9 eV decreases with Gd content. Increasing Gd content means decreasing Fe density of states below Fermi energy. This results in the suppression of Fe 3d to Gd 5d transition probability. On the contrary, the increase of Gd content is increasing the amplitude of the second Lorentz oscillator centered near 2.5 eV. Such behavior supports the assignment of the origin to Gd d-d transition. Our investigation of MO properties of  $\text{Gd}_x\text{Fe}_{(100-x)}$  showed that Gd substitution decreases both,  $\epsilon_{2r}$  and  $\epsilon_{2i}$  amplitudes. We attributed this to the fact that the magnetic moment of Fe is in this ferrimagnetic alloy stronger than the magnetic moment of Gd. Perpendicular anisotropy of  $\text{Gd}_x\text{Fe}_{(100-x)}$  was confirmed for all the samples. Moreover, we observed change in the magnetization direction to the opposite site when reaching the compensation concentration. We used one Dia transition to parameterize spectra of the off-diagonal elements of the  $\text{Gd}_x\text{Fe}_{(100-x)}$  permittivity tensor in terms of microscopic theory. We assumed that the MO effect

comes from different probabilities of transition between an orbital singlet ground state and split excited state and that Gd concentration decreases this splitting.

When investigating magnetic garnets, we started with Bix:YIGs thin films with various Bi concentrations and determined their full permittivity tensors at energies from 1.5 to 5 eV. TEM measurement confirmed that Bix:YIGs films grow uniformly and epitaxially on GGG. We found that bismuth substitution increases amplitudes of  $\epsilon_{1r}$  and  $\epsilon_{1i}$  in the measured spectral range. We observed optical transitions at 2.5, 3.2 and 4.4 eV and the absorption edge near 2.1 eV. As expected, we found that Bi substitution leads to the enhancement in MOKE and Faraday effects which is crucial for the garnet application potential. This result is also connected to the fact that bismuth increases  $\epsilon_{2r}$  and  $\epsilon_{2i}$  amplitudes at their extremes significantly. We used two Para transitions and three Dia transition to parameterize  $\epsilon_{2r}$  and  $\epsilon_{2i}$  in terms of microscopic theory. We found that Bi substitution increases amplitudes of almost all transitions. It however lowers energy of Dia transition at 4.65 eV which most likely exist due to charge transfers from oxygen to octahedral Fe. We associated strongest Dia transitions at 2.5 eV and 3.3 eV with transitions  $t_2(\text{Fe}^{3+}) \rightarrow t_2g(\text{Fe}^{2+})$  and  $eg(\text{Fe}^{3+}) \rightarrow e(\text{Fe}^{2+})$ . These are mainly responsible for the increase in MO effects. We attributed positive impact of Bi on these transitions to the increase in super-exchange interaction caused by the enhancement of electronic exchange.

As a second part of magnetic garnet research, we determined complete permittivity tensors of Bi1:NIGxGs and Bi2.5:NIGxGs thin films with different Ga concentrations at energies from 1.5 to 5.5 eV. We found that Ga substitution decreases amplitudes of  $\epsilon_{1r}$  for Bi1:NIGxGs below 4 eV and increasing them above. On the other hand, Ga substitution does not noticeably influence amplitudes of  $\epsilon_{1r}$  for Bi2.5:NIGxGs. We also found that Ga substitution increases absorption of Bi1:NIGxGs. However it decreases absorption of Bi2.5:NIGxGs. Furthermore, the absorption of Bi2.5:NIGxGs is almost 30% stronger than absorption of Bi1:NIGxGs. We attribute this to the higher Bi concentration. When looking at MO properties, we found that Bi substitution increases, and Ga substitution decreases amplitudes of MO effects. This is connected to  $\epsilon_{2r}$  and  $\epsilon_{2i}$  amplitudes which are increased by Bi and decreased by Ga substitution. To explain the effect of Ga properly we parameterized  $\epsilon_{2r}$  and  $\epsilon_{2i}$  spectra in terms of microscopic theory. We used four Para transitions to

represent crystal field transitions. We also used four Dia transitions. The main contribution comes from Dia transitions associated with transitions  $t_2(\text{Fe}^{3+}) \rightarrow t_2g(\text{Fe}^{2+})$  and  $e_g(\text{Fe}^{3+}) \rightarrow e(\text{Fe}^{2+})$ , mainly responsible for the remarkable increase in MO effects. We found that Ga substitution is decreasing these transitions. This is in accordance with the assumption, that Ga is mostly substituted for  $\text{Fe}^{3+}$  tetrahedral, which is crucial for both transitions. We also found that Ga substitution lowers energy of much smaller Dia transition at 4.65 eV which most likely exist due to charge transfers from oxygen to octahedral Fe. This is in accordance with the assumption that Ga is in a smaller percentage also substituted per  $\text{Fe}^{3+}$  octahedral.

Finally, we focused on the determination of full dielectric permittivity tensors of  $\text{CeHfCoO}$  thin films with different Hf concentrations in the spectral range from 1.5 to 5.5 eV. We found that Hf content decreases  $\epsilon_{1r}$  amplitudes in the whole measured spectral range. We also observed optical bandgap energies to be shifted from 3.21 to 4.1 eV when Hf content increased. We related these energies to  $\text{O}2p \rightarrow \text{Ce}4f$  electronic transitions. Similarly, we found that Hf doping decreases absorption in the whole measured spectral range. From this result we assumed that when is Hf replacing Ce in the material; it acts against optical vacancies from isolated  $\text{Ce}4f$  states localized within the optical bandgap, responsible for enhanced optical absorption. Absorption tail below 3.2 eV supported this theory and it was attributed to the effect of mid-gap defects. When investigating MO properties, we found that Faraday rotation extreme is shifting from 2.9 eV to higher energies. Rotation values decreased when Hf content increased. On the other hand, ellipticity values acted in the opposite manner. We also found that Hf substitution decreases both,  $\epsilon_{2i}$  and  $\epsilon_{2r}$  amplitudes and it is shifting their maxima to higher energies in the measure spectral range. This was explained by magnetoelastic effects which originate from distortions caused by in-plane compressive strain and vary with Ce-Co content. Hf substitution influences this content and reduces oxygen vacancies important for  $\text{CeHfCoO}$  magnetism. We parameterized  $\epsilon_{2r}$  and  $\epsilon_{2i}$  spectra in terms of microscopic theory by the sum of Para and Dia oscillator terms. We found that the main MO contribution comes from Dia transitions at (1.5-1.65 eV) which refers to localized 4f states in the band gap and at (3.75-4.3 eV) which refers to the oxygen electronic transitions.

## List of Abbreviations

**MO** - Magneto-optical

**SE** - Spectroscopic Ellipsometry

**MOKE** - Magneto-optical Kerr Effect

**EMA** - Effective Medium Approximation method

**MSA** – Multi Sample Analysis

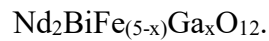
**MOD** - Metal Organic Decomposition

**MO-SLM** – Magneto-optical Spatial Light Modulator

**GGG** – Gadolinium Gallium Garnet,  $Gd_3Ga_5O_{12}$

**Bix:YIGs** – Bismuth substituted Yttrium Iron Garnets,  $Y_{3-x}Bi_xFe_5O_{12}$

**Bi1:NIGxGs** – Bismuth (1) and Gallium (x) substituted Neodymium Iron Garnets,



**Bi2.5:NIGxGs** - Bismuth (2.5) and Gallium (x) substituted Neodymium Iron



**CeHfCoO** – Hafnium and Cobalt substituted Cerium Oxide,  $Ce_{(0.95-x)}Hf_xCo_{0.05}O_{(2-\delta)}$



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