Fabrication and Characterization of $Y_3Fe_5O_{12}$|Pt|$Y_3Fe_5O_{12}$ Trilayers for Spin Current Based Experiments

Herstellung und Charakterisierung von $Y_3Fe_5O_{12}$|Pt|$Y_3Fe_5O_{12}$ Trilagen für Spinstrom basierte Experimente

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Chapter 1

Introduction

Today’s common electronic devices usually use the electrical properties of the electron. These devices reach physical limitations towards speed and miniaturization. Therefore, new methods have been actively researched in the last view years. One of them is the use of the electron spin leading to a new field called spin electronics or spintronics.

In this subject area new physical properties arise due to the development in thin film technologies. An example is the newly discovered spin magnetoresistance (SMR) in ferromagnetic insulating (FMI)/nonferromagnetic (NM) hybrid structures, so called bilayers [1]. This new magnetoresistance (MR) is different to the known anisotropic [2], giant [3, 4] and tunnel [5, 6] magnetoresistance since the electrical current need not pass through a magnet [1]. Here the MR is induced in the NM layer by a nonequilibrium proximity effect [1], which depends on the magnetic properties of the FMI layer although the charge current is not able to pass through the magnet [7]. The SMR effect results of a spin current that can flow from the NM into the FMI layer depending on the magnetization direction of the FMI thin film. This spin current is caused by the spin Hall effect (SHE) in the NM layer [8] while the spin transfer into the FMI is caused by spin scattering at the NM/FMI interface [7]. When the spin polarization of the spin current is perpendicular to the FMI magnetization the spins can pass through the spin-transfer torque effect [9] into the FMI.

The SMR effect has already been quantitatively studied for bilayers in Ref. [10]. However, the MR effect is about 0.01% being much smaller compared to the other mentioned MRs [7]. Anyway, the SMR effect allows to sense the magnetization in the FMI layer over the SHE and inverse SHE [11, 12] and makes the integration of insulating ferromagnets into electronic circuits possible [1]. A potential application is a so called magnetoresistive random access memory (MRAM) being a non-volatile memory device in contrast to common static and dynamic random access memories [13].

The small change in the resistance of the NM layer is expected to be more than twice larger in FMI|NM|FMI trilayers compared to bilayers when the thickness of the NM layer is comparable with the spin diffusion length [14]. Furthermore, a trilayer has an additional degree of freedom in the magnetization orientation of the second FMI layer. Therefore, so called spin valves realize three different resistance values for the NM layer when a current is applied through it parallel to the FMI interfaces [14]. This could lead to a higher storage density in MRAMs being based on ternary instead of binary logic.

For the fabrication of a trilayer the following materials are used. The FMI layer is realized with the ferrimagnetic electrical insulator $Y_3Fe_5O_{12}$ (YIG). This material is already well known for applications in high frequency technology due to its small spectral linewidth of about 0.02 mT in the ferromagnetic resonance [15]. Furthermore, YIG shows excellent magneto-optical properties [16]. YIG can be epitaxially grown using pulsed laser deposition (PLD) [17–19]. As substrates gadolinium gallium garnet $Gd_3Ga_5O_{12}$ (GGG) and yttrium aluminium garnet $Y_3Al_5O_{12}$ (YAG) are used. For the NM spacer platinum (Pt) is used because of its high spin-orbit coupling [20], which is essential for the creation of spin currents over the SHE. Pt is
Chapter 1 Introduction

deposited using electron beam evaporation (EVAP). The complete fabrication of a YIG|Pt|YIG trilayer is realized in an ultra high vacuum (UHV) cluster.

This thesis is organized as follows. Chapter 2 describes the basic theoretical concepts of the SMR effect and how it can be measured in a bilayer. Furthermore, the SMR effect of a bilayer is compared to the theoretical expected one of a trilayer. Moreover, spin valves are briefly explained in this chapter. Finally, some basic properties of the used materials for the FMI and NM layers as well of the substrates are explained. Chapter 3 deals with the fabrication of a trilayer on the substrate GGG. To this end the fabrication steps are divided into several production stages for a better analysis of the grown thin films using x-ray diffractometry, atomic force microscopy and superconducting quantum interference device (SQUID) magnetometry. The sample fabrication and the different analysis methods are described in more detail in appendix A. Chapter 4 covers the trilayer fabrication on the second substrate YAG. In chapter 5 the achieved results are summarized concerning the goal of a successful trilayer fabrication with the described properties. Additionally an outlook is given about further investigations.
Chapter 2

Theoretical foundations

In the first part of this chapter some theoretical basics of the spin Hall magnetoresistance (SMR) are described and how it can be measured using a patterned Hall bar geometry of a ferromagnetic insulating (FMI)/nonferromagnetic (NM) hybrid structure. Furthermore, the SMR effect of a FMI|NM bilayer is compared to the one of a FMI|NM|FMI trilayer and the application of trilayers as spin valves is explained. The second part deals with the materials and their properties which are used for the fabrication of a FMI|NM|FMI trilayer.

2.1 Spin Hall magnetoresistance and spin valve

The fabrication of a trilayer is the next milestone for the study of ferromagnetic insulating (FMI)/nonferromagnetic (NM) thin film systems. So called bilayers consisting of a FMI and NM layer have already been quantitatively studied in Ref. [10] for the examination of new effects as the spin magnetoresistance (SMR) [1]. Figure 2.1(a) shows such a bilayer.

\[ J_{s,\text{abs}} = \alpha_{\text{SH}} \left( -\frac{\hbar}{2e} \right) J_q \times s \]  

(a) Bilayer consisting of a ferromagnetic insulator (FMI)/nonferromagnetic (NM) hybrid structure (based on Ref. [7]). A charge current density \( J_q \) in the NM induces over the spin Hall effect a spin current \( J_s \) depending on the spin polarization \( s \) (indicated by arrows going through the electrons). These spins are accumulated at the NM|FMI interface and can be absorbed over the spin torque effect into the FMI (\( J_{q,\text{abs}} \)) if the magnetization direction \( M \) is perpendicular to \( s \). (b) Patterned Hall bar geometry of a FMI|NM bilayer grown on a substrate (from Ref. [10]). The longitudinal resistance is measured over \( \rho_{\text{long}} = V_{\text{long}} w t_{\text{NM}} / I_q l \) with the length \( l \), the width \( w \) and the thickness \( t_{\text{NM}} \) of the NM layer as well as the applied current \( I_q \) and the measured longitudinal voltage drop \( V_{\text{long}} \). Furthermore, the right-handed coordinate system is spanned by the three unit vectors \( j \), \( t \) and \( n \).

A charge current density \( J_q \) in the NM layer induces over strong spin-orbit coupling a spin current density \( J_s \) due to the spin Hall effect [8]. According to Ref. [10] both are related by
with the spin polarization $s$ (arrows of electrons in Fig. 2.1(a)), the positive elementary charge $e$ and the spin Hall angle $\alpha_{\text{SH}} = \sigma_{\text{SH}} / \sigma$ (ratio of spin Hall conductivity $\sigma_{\text{SH}}$ and electric conductivity $\sigma$). Without a FMI attached to the NM layer the spin current traveling to the film surface is reflected back into the NM and converted into an electrical current over the inverse spin Hall effect [1]. This behavior of the spin current $J_s$ can be influenced in a FMI bilayer as shown in Fig. 2.1(a) over the magnetization direction $M$ of the FMI. When the magnetization $M$ is perpendicular to the spin polarization $s$ the spin reflection is suppressed and the spin current is absorbed ($J_{s,\text{abs}}$) even at the interface to a magnetic insulator over the spin torque effect [9]. This effect allows the transfer of angular momentum and leads to a spin flip in the NM layer [1, 10]. On the other hand, the reflection is maximized when $M$ is collinear to $s$ since no angular momentum transfer is possible. In this simple picture a higher resistance of the NM layer is expected if the spin current is absorbed at the FMI/NM interface and a lower if not.

The dependence of the resistance in the NM layer on the magnetization direction of the FMI layer can be measured by patterning a Hall bar geometry in the bilayer as shown in Fig. 2.1(b).

![Figure 2.2](image)

**Figure 2.2:** (a) Dependence of the spin magnetoresistance (SMR) effect $-\rho_1/\rho_0$ on the nonferromagnetic (NM) layer thickness $t_{\text{NM}}$. Additionally the theoretical expected SMR effect of a trilayer is illustrated which is greater than two times the curve of the bilayer [14]. (b) Principal of a spin valve using the SMR effect. In the left spin valve (consisting of a FMI|NM|FMI trilayer grown on a substrate) both magnetizations $M_1$ and $M_2$ of the ferromagnetic insulating (FMI) layers are perpendicular to the spin polarization of the spin current which is induced by the charge current density $J_q$. As described in Fig. 2.1(a) the spin current can be absorbed by the FMI leading to a higher longitudinal resistance in the NM layer. In the right spin valve the upper magnetization $M_1$ is turned parallel to the spin polarization resulting in a lower resistance state in the NM layer.

According to Ref. [10] the longitudinal resistance $\rho_{\text{long}} = V_{\text{long}} w t_{\text{NM}} / (I_q l)$ of the NM layer can be parametrized as

$$\rho_{\text{long}} = \rho_0 + \rho_1 m_t^2$$

(2.2)

with $m_t$ as the projection of the magnetization $m = M/M_{\text{saturation}}$ on the coordinate vector $t$, the proportionality factor $\rho_1 < 0$ and $\rho_0$ as the expected resistance for $m_t = 0$. The ratio $|\rho_1/\rho_0| = -\rho_1/\rho_0$ is the SMR effect which is plotted in Fig. 2.2(a) over the thickness $t_{\text{NM}}$ of the NM layer based on the theoretical calculations in Ref. [14]. When $t_{\text{NM}}$ is comparable to the spin-diffusion length $\lambda$ in the NM (maximal possible spin accumulation) a maximum occurs in the ratio $|\rho_1/\rho_0|$. Rotating the magnetization $M$ in the FMI at this maximal ratio leads to the highest resistance change in the NM layer according to Eq. (2.2) ($\cos^2$-dependence of $M$ for an in-plane rotation parallel to the NM|FMI interface). Furthermore, the SMR effect differs from the anisotropic magnetoresistance [2] which is quantitatively shown in [10].
As illustrated in Fig. 2.2(a) the SMR effect in a FMI|NM|FMI trilayer is expected to be more than twice as large as in a FMI|NM bilayer according to Ref. [14]. This should result in better measurable resistance changes of the NM layer. The increase of the SMR effect is caused by the additional FMI layer which allows absorption and reflection of the spin current at two interfaces depending on the magnetization direction in both FMI layers.

The existence of two FMI layers with adjustable magnetization orientations \( \mathbf{M}_1 \) and \( \mathbf{M}_2 \) in FMI|NM|FMI trilayers enables the possibility for magnetization dynamics [14] in so called spin valves as shown in Fig. 2.2(b). The charge current flows as depicted by the blue arrow. According to Eq. (2.1) a spin current density is induced. In the left spin valve both magnetizations \( \mathbf{M}_1 \) and \( \mathbf{M}_2 \) of the FMI layers are orientated perpendicular towards the spin polarization in the NM [10] leading to a higher resistance of the NM thin film. In the right spin valve the magnetization of the upper FMI layer \( \mathbf{M}_1 \) is collinear to the spin polarization resulting in a lower resistance of the NM layer since no spin absorption into the upper FMI layer is possible. When \( \mathbf{M}_2 \) is also rotated into a collinear orientation a third resistance state with the lowest value can be realized.

The described theoretical properties highly motivate the fabrication of a FMI|NM|FMI trilayer for an experimental review. Therefore, the next section describes some basic properties of the used materials for the FMI and NM layers as well as of the substrates which are used.

### 2.2 Material properties

For the ferromagnetic insulator (FMI) synthetic yttrium iron garnet \( \text{Y}_3\text{Fe}_5\text{O}_{12} \) (YIG) is used. Its crystal structure is shown in Fig. 2.3.

![YIG Crystal Structure](image)

**Figure 2.3:** \( \text{Y}_3\text{Fe}_5\text{O}_{12} \) (YIG) crystal structure (taken from Ref. [21]). The cubic unit cell contains yttrium (Y\(^{3+}\), 24(c) positions), iron (Fe\(^{3+}\), 16(a) and 24(d) positions) and oxygen (O\(^{2-}\), 96(h) positions) ions [22]. These mentioned positions are the so called Wyckoff positions [23]. Overall there are 160 atoms in one unit cell which corresponds to 8 molecular formulas of YIG. The Y\(^{3+}\) cations are decahedral coordinated (red shaded) while the Fe\(^{3+}\) cations are tetrahedral and octahedral coordinated (gray and black shaded).

While the three Y\(^{3+}\)-ions are decahedral coordinated two Fe\(^{3+}\)-ions are octahedral and three Fe\(^{3+}\)-ions are tetrahedral coordinated (cf. Fig. 2.3). Due to an indirect superexchange coupling [24] of the Fe\(^{3+}\)-cations over the O\(^{2-}\)-ions, the two Fe\(^{3+}\) coordinations have an antiparallel spin orientation. Therefore, two magnetic sublattices exist in YIG which compensate the magnetic moment of four Fe\(^{3+}\)-ions. Thus, the total magnetization is just caused by one Fe\(^{3+}\)-ion leading to a ferrimagnetic ordering in YIG. The Y\(^{3+}\) and O\(^{2-}\) ions are diamagnetic and do not account for the overall magnetization because of completed electron shells.

At 300 K YIG has a saturation magnetization of 141.65 kA m\(^{-1}\) [25]. Due to the high Curie temperature of 559 K [25] YIG still has a high remanence at room temperature. For temperatures larger than the Curie temperature YIG gets paramagnetic. The coercive field is extremely small with values below 15 mT which highly depends on the growth conditions during the fabrication process [10].
Chapter 2 Theoretical foundations

For the growth of YIG two different (111)-orientated substrates from CrysTec [26] are used. Gadolinium gallium garnet $\text{Gd}_3\text{Ga}_5\text{O}_{12}$ (GGG) also crystallizes with eight molecular formulas per cubic unit cell. Due to a missing superexchange coupling GGG is paramagnetic which is caused by the Gd$^{3+}$-ions. Furthermore, yttrium aluminium garnet $\text{Y}_3\text{Al}_5\text{O}_{12}$ (YAG) is used which crystallizes as YIG. Since all appearing ions have completed electron shells, YAG is diamagnetic.

As nonferromagnetic (NM) material paramagnetic platinum (Pt) is used which has a strong spin-orbit coupling interaction [20]. This is relevant for the creation of pure spin currents as described in Sec. 2.1.

<table>
<thead>
<tr>
<th>Material</th>
<th>YIG</th>
<th>GGG</th>
<th>YAG</th>
<th>Pt</th>
</tr>
</thead>
</table>

Table 2.1: Lattice constants of YIG, GGG, YAG and Pt.

Table 2.1 shows the lattice constants of the four materials YIG, GGG, YAG and Pt. The relevant lattice mismatches $\varepsilon$ between these materials are calculated by

$$\varepsilon = \left| \frac{a_{\text{film}} - a_{\text{substrate}}}{a_{\text{substrate}}} \right|$$

and the results are listed in Tab. 2.2. Therefore, nine unit cells of Pt are assumed to grow on one unit cell of GGG, YAG or YIG (which is treated as the substrate in this case) in the corresponding columns.

<table>
<thead>
<tr>
<th>Material</th>
<th>GGG</th>
<th>YIG</th>
<th>GGG</th>
<th>Pt</th>
<th>YAG</th>
<th>YIG</th>
<th>YAG</th>
<th>Pt</th>
<th>YIG</th>
<th>Pt</th>
</tr>
</thead>
<tbody>
<tr>
<td>Lattice mismatch</td>
<td>0.03 %</td>
<td>5.00 %</td>
<td>3.13 %</td>
<td>2.00 %</td>
<td>4.98 %</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Table 2.2: Relevant lattice mismatches between YIG, GGG, YAG and Pt.

The next chapter starts with the fabrication of a YIG|Pt|YIG trilayer on the substrate GGG because of the smaller lattice mismatch between GGG and YIG compared to YAG and YIG (cf. Tab. 2.2). This smaller lattice mismatch is important for the epitaxial growth of a high quality crystalline YIG thin film using pulsed laser deposition.
Chapter 3

\( \text{Y}_3\text{Fe}_5\text{O}_{12}|\text{Pt}|\text{Y}_3\text{Fe}_5\text{O}_{12} \) trilayer on \( \text{Gd}_3\text{Ga}_5\text{O}_{12} \) substrate

In this chapter the fabrication of a YIG|Pt|YIG trilayer on a (111)-orientated GGG substrate is described. Therefore, the next section explains the used fabrication and analysis methods. The succeeding sections discuss the different fabricated sample types towards their structural, surface and magnetic properties to optimize each fabrication stage and, thus, to find the best growth parameters for a YIG|Pt|YIG trilayer.

3.1 Fabrication and analysis methods

The fabrication of YIG|Pt|YIG trilayers on (111)-orientated GGG substrates is divided into several steps as shown in Fig. 3.1. In this way the influences of each new deposited layer of YIG or Pt can be examined to optimize the corresponding fabrication stage.

![Figure 3.1](image)

*Figure 3.1: (a) YIG|Pt|YIG trilayer on a (111)-orientated GGG substrate. (b)–(e) Different sample types that are fabricated to examine and optimize the various thin film systems. (d) is also referred to as inverse bilayer and (e) as bilayer.*

The YIG thin films are epitaxially grown by pulsed laser deposition (PLD) from a stoichiometric polycrystalline YIG target. The Pt layers are deposited *in situ* (without breaking the vacuum) using electron beam evaporation (EVAP).

For all samples the structural and surface properties are investigated using x-ray diffractometry (XRD) and atomic force microscopy (AFM). The magnetic properties of the samples containing YIG are determined by superconducting quantum interference device (SQUID) magnetometry. Both fabrication methods, PLD and EVAP, as well as the three analysis methods XRD, AFM and SQUID are described in more detail in appendix A.

The next section discusses the optimization process of the YIG deposition on (111)-orientated GGG substrates as depicted in Fig. 3.1(b).
3.2 $Y_3Fe_5O_{12}$ thin films on $Gd_3Ga_5O_{12}$

3.2.1 Growth parameters

The optimal set of YIG growth parameters are a substrate temperature of $T_S = 550^\circ$C during deposition, an energy density of $\rho_L = 2\, \text{J/cm}^2$ at the target, a repetition rate of $f = 10\, \text{Hz}$ of the KrF excimer laser and an oxygen atmosphere of $p = 25\, \mu\text{bar}$ (cf. Ref. [10]). Since the second YIG film of the trilayer (cf. YIG2 in Fig. 3.1(a)) is grown on Pt, an oxygen atmosphere can not be used because Pt oxidizes immediately. Furthermore, a trilayer has already been fabricated with the parameters from above but no conductivity of the Pt thin film was found. This indicates the formation of the insulating oxide PtO$_x$ (increasing isolation with $x$ [32, 33]). Therefore, an argon atmosphere (inert gas) is chosen to avoid the oxidation of Pt.

For the growth optimization of YIG in an argon atmosphere a substrate temperature $T_S$ of $550^\circ$C, $450^\circ$C and $350^\circ$C is used while the pressure is kept constant at $p = 25\, \mu\text{bar}$. All the other parameters are unchanged. The structural, surface and magnetic properties of these YIG thin films are discussed in the following sections.

3.2.2 X-ray diffractometry

$2\theta - \omega$ scans and out-of-plane lattice constants

Figure 3.2(a) shows a complete $2\theta - \omega$ scan with the allowed reflections of the garnet structure in the (111) direction. Only the GGG substrate and YIG thin film reflexes are visible lying very close to each other due to the small lattice mismatch (see Sec. 2.2). The $2\theta - \omega$ scans of GGG|YIG samples grown at three different substrate temperatures in an argon atmosphere as well as a reference sample grown with the optimal parameters in oxygen are shown in Fig. 3.2(b) and (c). The existence of YIG reflections indicates crystalline growth of the thin films. Furthermore, Fig. 3.2(b) shows satellites due to Laue oscillations, which is evidence for a coherent growth of the YIG lattice planes. Both oscillations marked with an asterisk (*) have a too high intensity $I$ for pure Laue oscillations and are caused by impurity phases that seem to grow at high temperatures ($T_S \geq 550^\circ$C) in an argon atmosphere.

Figure 3.2(d) displays the calculated out-of-plane lattice constants $a_{YIG}$ from all four samples using the Bragg equation (A.1) and assuming an undisturbed cubic lattice symmetry of YIG as described by Eq. (A.2). For this purpose the YIG (444) and YIG (888) reflexes marked with an arrow in Fig. 3.2(b) and (c) are fitted with a Gaussian function to extract the $2\theta$-value of the maximum and its statistical error $\Delta(2\theta)$. The errors shown in Fig. 3.2(d) are calculated using Gaussian error propagation on Eq. (A.1) according to Ref. [34]:

$$\Delta a_{YIG} = \Delta(2\theta) \cdot \frac{a_{YIG}}{2 \cdot \cot\left(\frac{(2\theta)}{2}\right)} \tag{3.1}$$

The resulting lattice constants calculated from the YIG (444) (cf. solid squares in Fig. 3.2(d)) and YIG (888) (cf. open circles in Fig. 3.2(d)) reflections, match within the error bar for each sample. The YIG thin film grown in oxygen has a lattice constant lying close to the literature value of bulk YIG [27, 28]. For the temperature series in argon the YIG thin film grown at the lowest substrate temperature of $350^\circ$C has the smallest difference to the literature value while the other two thin films show the trend of a raising lattice constant with increasing substrate temperature $T_S$ (indicated by the black line in Fig. 3.2(d)). This can be explained by an increase of oxygen vacancies in the crystal structure of YIG with an increasing substrate temperature leading to a volume enlargement of the unit cell. Using an oxygen atmosphere prevents these vacancies as confirmed by the reference sample compared to the one grown at $550^\circ$C in argon.
Figure 3.2: (a) Complete $2\theta - \omega$ scan of one GGG|YIG sample grown in argon at a substrate temperature $T_S$ of 550°C. (b) and (c) Enlargement around the substrate reflections GGG (444) and (888) for the temperature series in argon and the reference sample grown in oxygen. The arrows mark the $2\theta$ values of the YIG (444) and (888) reflections that are used to calculate the out-of-plane lattice constants $a_{YIG}$ in (d) from the Bragg equation assuming a cubic symmetry of YIG. The arrows in (b) label the detector position for the measured rocking curves in Fig. 3.3(a) except for oxygen. Here the orange arrow marks the $2\theta$-value since the YIG (444) reflection nearly overlaps with the substrate peak. The asterisks (*) denote satellites which have a too high intensity $I$ to be pure Laue oscillations and are caused by impurity phases in the YIG thin film.

The arrows marking the YIG (444) reflections in Fig. 3.2(b) are also the detector positions for the measured rocking curves (next paragraph) except for the sample grown in oxygen. Here the orange arrow labels the $2\theta$-value since the YIG (444) peak nearly overlaps with the substrate reflection due to the small lattice mismatch between GGG and YIG (cf. Tab. 2.2).

**Rocking curves and full width at half maximums**

The measured rocking curves at the $2\theta$-positions marked in Fig. 3.2(b) are shown in Fig. 3.3(a) and the extracted full width at half maximums (FWHM) in Fig. 3.3(b) using Gaussian fits. The FWHM is a quantity for the mosaic spread of the lattice planes with a low one indicating a good crystalline quality [35]. All narrow components of the rocking curves have a FWHM lower than 0.04° revealing an excellent crystalline quality of the YIG thin films independent of the used gas atmosphere during the growth (argon or oxygen). The broader components with lower intensities have larger FWHM values and, thus, higher mosaic spreads (upper part in Fig. 3.3(b)). This is probably caused by the first lattice planes growing on the substrate GGG.
Figure 3.3: (a) Measured rocking curves from the YIG (444) reflexes as mentioned in Fig. 3.2. For a better comparison the $\omega$-value of the maximum of each peak $\omega_{\text{max}}$ is determined with a Gaussian fit and subtracted from the $\omega$-values leading to $\Delta \omega = \omega - \omega_{\text{max}}$. (b) Extracted full width at half maximums (FWHM) from the Gaussian fits which are plotted versus the corresponding substrate temperature $T_S$. The lower part of the plot contains the FWHMs of the narrow components (solid squares) while the upper part shows the broader components (open circles). The sketched errors are calculated over two times the statistical error received from the Gaussian fits.

before the coherent growth begins. The portion of these tilted lattice planes is very low as shown by the intensity relation between the corresponding peak components. The overall thickness and surface roughness of the grown YIG thin films can be determined by x-ray reflectometries.

Reflectometries, thicknesses and roughnesses

The measured reflectometries are shown in Fig. 3.4(a) and the extracted thicknesses and roughnesses using the simulation software LEPTOS (see Sec. A.2.1) are depicted in Fig. 3.4(b). These simulations are shown in Fig. 3.5(a)–(d) for each thin film.

Figure 3.4: (a) Reflectometries with thickness fringes caused by the YIG thin films. (b) Extracted thicknesses $d_{YIG}$ and roughnesses (open circles) for the YIG thin films (solid squares). The roughness $R_{\text{RMS}}$ is the root mean square (RMS) value of the average surface height calculated with the simulation software, which also allows the determination of the drawn errors. Both thickness and roughness are plotted versus the substrate temperature $T_S$. 

The reflectometries in Fig. 3.4(a) show Kiessig fringes [36] due to the YIG thin films. These oscillations vanish very fast for the sample grown in argon at a substrate temperature of 550 °C. This might be caused by a high substrate surface roughness (cf. Fig. 3.4(b)) or the mentioned impurity phases, which might also be responsible for the difference between simulation and measurement in Fig. 3.5(a). For the other thin films the corresponding simulation and measurement agree perfectly as apparent from Fig. 3.5(b)–(d).

The different thicknesses are not a suitable quantity for comparison because the thickness is highly dependent on many parameters during growth. Even YIG thin films grown with nominal the same deposition parameters exhibit different thicknesses (see the sample view in Tab. B.1). Also the determined roughnesses have to be considered carefully since they are representing a line shaped profile as described in Sec. A.2.1. Here the atomic force microscopy (AFM) provides a much better insight into the surface topography and hence into the roughness which is discussed in the next section. According to Fig. 3.4(b) the reference sample has the lowest surface roughness while only changing the gas atmosphere from oxygen to argon results in an increase of the roughness of about eight times.
3.2.3 Atomic force microscopy

The surface topographies measured with the AFM of the three thin films grown in argon and the reference sample grown in oxygen are shown in Fig. 3.6.

![Surface topographies measured with an atomic force microscope using the tapping mode as described in Sec. A.2.2. The scan size of each picture is 5µm × 5µm as labeled by X and Y. The Z value is the maximal peak to valley value that exists in the picture. The surface roughness \( R_{\text{RMS}} \) (root mean square (RMS)) is calculated over the average height of the surface according to Ref. [34]. As obvious from (a), (b) and (d) facets seem to form for substrate temperatures \( T_S \geq 450 \degree \text{C} \) independent of the gas atmosphere that is used during the growth. These facets are missing in (c) leading to a flat but rough surface.](image)

The roughnesses from Fig. 3.6(a) and (c) match to the one determined by the x-ray reflectometry in Fig. 3.4(b). This is not the case for Fig. 3.6(b) and (d) where a much higher surface roughness is detected especially for the reference sample. For substrate temperatures \( T_S \geq 450 \degree \text{C} \) facets seem to form as observable in Fig. 3.6(a), (b) and (d) independent of the chosen gas atmosphere during growth. This behavior of the thin films is discussed in detail in Sec. 3.7. The next section describes the magnetic properties of the YIG thin films.
3.2.4 SQUID magnetometry

The in-plane magnetization $M$ of the YIG thin films is measured at room temperature (300 K) using SQUID magnetometry as described in Sec. A.8. The resulting hysteresis curves are shown in Fig. 3.7(a). The subtraction of the magnetic background from the paramagnetic substrate GGG causes the splitting of the data points in the saturation magnetization area and a small non-centered hysteresis curve around the origin.

![Figure 3.7](image)

Figure 3.7: (a) Measured magnetic hysteresis of the YIG thin films at 300 K. The magnetization $M$ is plotted versus the external field $\mu_0 H$ which is varied from 100 mT to $-100$ mT and back again. (b) Saturation magnetization $M_S$ (solid squares) and coercive field $\mu_0 H_C$ (open circles). Both quantities are plotted versus the corresponding substrate temperature $T_S$.

The data points between $-10$ mT and $-100$ mT as well as between 10 mT and 100 mT are fitted with a horizontal line to receive two saturation magnetization $M_{S,\text{left}}$ and $M_{S,\text{right}}$ with a corresponding error $\Delta M_{S,\text{left}}$ and $\Delta M_{S,\text{right}}$. This error is calculated over the maximal distance in the $y$ direction from the data points in the corresponding regions. Finally the magnetization $M_{S,YIG}$ of the YIG thin film is calculated to

$$M_{S,YIG} = \frac{M_{S,\text{left}} + M_{S,\text{right}}}{2} \quad \text{with} \quad \Delta M_{S,YIG} = \frac{1}{2} \sqrt{(\Delta M_{S,\text{left}})^2 + (\Delta M_{S,\text{right}})^2}. \quad (3.2)$$

The resulting saturation magnetizations and their corresponding uncertainties are shown in Fig. 3.7(b). Since the measured magnetic moment $m$ has to be divided by the thin film volume $V$ to receive the magnetization $M$ (cf. Eq. (A.3)) an additional error from the volume determination according to Eq. (A.4) has to be added. Assuming a thin film surface area of $A \approx 23 \text{ mm}^2$ with an error of $\Delta A \approx 0.1 \text{ mm}^2$ and a thickness of $d \approx 50 \text{ nm}$ with an uncertainty of $\Delta d \approx 0.5 \text{ nm}$ the error can be estimated by

$$\Delta M = M \cdot \sqrt{\left(\frac{\Delta A}{A}\right)^2 + \left(\frac{\Delta d}{d}\right)^2} \approx 1.5 \text{ kA m}^{-1} \quad (3.3)$$

using Gaussian error propagation on Eq. (A.3) and Eq. (A.4) according to Ref. [34]. Thereby, the literature value for the saturation magnetization of bulk YIG $M_S = 141.65 \text{ kA m}^{-1}$ [25] is substituted for the magnetization $M$. However, this error can be neglected compared to the one resulting from Eq. (3.2). Figure 3.7(b) also contains the determined coercive fields $\mu_0 H_C$.

As expected these coercive fields with $\mu_0 H_C < 3 \text{ mT}$ are very small and confirm the described magnetic property of YIG in Sec. 2.2.
The calculated saturation magnetizations from Eq. (3.2) match within their uncertainties for all thin films grown in argon at three different substrate temperatures as well with the reference sample. This is surprising since the mentioned oxygen vacancies in Sec. 3.2.2 should decrease the magnetic moment in YIG at 300 K due to a weaker Fe$^{3+}$-O$^{2-}$-Fe$^{3+}$ superexchange interaction in the unit cell [27, 28]. Here measurements of the Curie temperature could give a better insight. Only the sample grown in argon at 550°C has a lower saturation magnetization compared to the other two thin films grown in argon which might be caused by the impurity phases.

### 3.2.5 Conclusion

The examination of the structural, surface and magnetic properties of YIG thin films grown in argon clearly provides an adequate temperature for the fabrication of YIG thin films for YIG|Pt|YIG trilayers: From the XRD measurements a substrate temperature $T_S < 550{\degree}C$ is appropriate since for $T_S = 350{\degree}C$ and $T_S = 450{\degree}C$ no formation of impurity phases is detected. Furthermore, these thin films have less oxygen vacancies and a better crystalline quality compared to 550°C. This trend is also confirmed by SQUID magnetometry measurements where a slightly higher saturation magnetization is observed. Finally AFM measurements confirm a substrate temperature of $T_S = 350{\degree}C$ since for YIG thin films fabricated at 350°C no facets are formed, which is important for the deposition of Pt in the next fabrication step of the trilayer. Only a flat surface of the YIG thin film can be completely coated by a Pt layer. These facets are the reason for the decreasing substrate temperatures mentioned in Sec. 3.2.1. Furthermore, the AFM result of the reference sample shows that even the standard parameters for an oxygen atmosphere can not be used to grow the first YIG layer of the trilayer (cf. YIG1 in Fig. 3.1(a)) due to the formation of facets as mentioned before. This is an additional motivation to use an argon atmosphere for the YIG thin film growth.

### 3.3 Pt thin films on Gd$_3$Ga$_5$O$_{12}$

The next fabrication step of the trilayer is the deposition of a Pt layer in the EVAP chamber (cf. Fig. 3.1(c)). Since the Pt layer is subsequently reheated to grow the second YIG layer on top it is mandatory to examine the behavior of Pt towards different substrate temperatures in the reheating process. For this purpose a Pt thickness of 10 nm is set in the EVAP process and afterwards the sample is transferred without breaking the vacuum into the PLD chamber. There it is annealed for one hour in an argon atmosphere with a pressure of 25 µbar at a substrate temperatures of $T_S = 550{\degree}C$, 450°C or 350°C. The next section describes the structural properties of the annealed Pt thin films using x-ray diffractometry.

#### 3.3.1 X-ray diffractometry

$2\theta - \omega$ scans and rocking curves

The three $2\theta - \omega$ scans of the Pt thin films on GGG substrates annealed at three different substrate temperatures are shown in Fig. 3.8(a). This plot reveals a significant change compared to the results in Ref. [10] where Pt is deposited in the EVAP but not annealed afterwards. There no Pt peak is visible in the $2\theta - \omega$ scan since Pt grows polycrystalline in the EVAP. In Fig. 3.8(a) a clear Pt (111) reflex is visible especially for an annealing temperature of 550°C. Fitting this reflection with a Gaussian curve and using Eq. (A.1) and Eq. (3.1) as described in Sec. 3.2.2 results in a lattice constant of $a_{Pt} = (0.3914\pm0.0016)\text{nm}$, which is in agreement
3.3 Pt thin films on Gd₃Ga₅O₁₂

![Graph](image)

**Figure 3.8:** (a) $2\theta - \omega$ scans of Pt thin films on GGG annealed for one hour at three different substrate temperatures in an argon atmosphere with a pressure of $p_{\text{Ar}} = 25 \, \mu\text{bar}$. (b) Measured rocking curves from the Pt (111) peaks which are shifted ($\Delta \omega$) as described in Fig. 3.3 for a better comparison. Furthermore, the Gaussian fit is shown for the black curve to determine the FWHM.

with the literature value of $a_{\text{Pt}} = 0.3920\, \text{nm}$ [31]. The Pt (111) reflections for $T_S = 350\, ^\circ\text{C}$ and $450\, ^\circ\text{C}$ can not be well fitted and are hence not evaluated.

The occurrence of a Pt reflection can be explained by growing crystallites in the initially polycrystalline Pt layer. The Debye-Scherrer formula [37] implies that the crystallite size in a polycrystalline material is inversely proportional to the FWHM of the corresponding peak in a $2\theta - \omega$ scan. Thus, during the annealing at $550\, ^\circ\text{C}$ the largest crystallites grow along the (111) direction due to the smallest FWHM of the corresponding Pt peak. This growing direction is the distinguished one from Pt based on the face centered cubic unit cell [31].

Figure 3.3(b) shows the measured rocking curves from the Pt (111) reflections. The FWHM for the Pt thin film annealed at $550\, ^\circ\text{C}$ reveals a very high mosaic spread denoting that the Pt thin film is not single crystalline. For the other rocking curve representing the annealing at $450\, ^\circ\text{C}$ and $350\, ^\circ\text{C}$ an infinite FWHM reveals that Pt is still polycrystalline.

**Reflectometries and simulations**

The measured reflectometries of the annealed Pt thin films are shown in Fig. 3.9(a). To determine the thickness of the Pt thin films without annealing a reflectometry is directly measured after the deposition in the EVAP (cf. blue line in Fig. 3.9(a)). The simulation of this reflectometry is shown in Fig. 3.9(b) leading to a Pt thickness of $d_{\text{Pt}} = (6.9 \pm 0.1)\, \text{nm}$. The reason for the different thickness compared to the set one of $10\, \text{nm}$ is explained in Sec. A.1.3. After this step the sample is reintroduced into the UHV cluster and annealed at $550\, ^\circ\text{C}$. The reflectometry measured afterwards (cf. black curve in Fig. 3.9(a)) reveals no Pt fringes. The same applies for an annealing temperature of $450\, ^\circ\text{C}$. Furthermore, both reflectometries have a loss in the intensity $I$ of one magnitude for small $2\theta$ angles indicating diffuse scattering. Only for an annealing temperature of $350\, ^\circ\text{C}$ the Pt fringes are still visible. The simulation of this curve in Fig. 3.9(b) leads to a Pt thickness of $d_{\text{Pt}} = (6.2 \pm 0.1)\, \text{nm}$ which is smaller than the deposited thickness in the EVAP determined by the simulation of the blue curve. This difference could be caused by measurement uncertainties (EVAP and XRD) or a beginning chemical transformation of the Pt thin film through the annealing process.

The vanishing Pt oscillations for an annealing temperature of $550\, ^\circ\text{C}$ or $450\, ^\circ\text{C}$ could be caused by Pt that diffuses into the substrate GGG leading to a rough interface. Furthermore, Pt could vaporize due to its small thickness and the low pressure of $25 \, \mu\text{bar}$ in the PLD chamber.
Another possibility is the reaction of Pt with the elements of GGG or the beginning crystallization, which is highly temperature dependent according to the $2\theta - \omega$ scan in Fig. 3.8(a). For a more detailed discussion further experiments are necessary as reflection high-energy electron diffraction (RHEED) [38, 39] during the annealing process or element specific investigations. In addition, it is not possible to simulate the two reflectometries in Fig. 3.9(a) assuming the ideal system GGG|Pt.

The determined roughnesses in Fig. 3.9(b) show that the deposited Pt layer before and after the annealing at $350^\circ$C should lead to a very flat surface. This is examined in more detail in the next section by AFM measurements.

### 3.3.2 Atomic force microscopy

The measured surface topographies of the annealed Pt thin films using AFM are shown in Fig. 3.10. Figure 3.10(b) and (c) reveal that also a Pt thin film shows facets on a GGG substrate for annealing temperatures equal or greater than $450^\circ$C. These facets could be the reason for the intensity loss of the reflectometry in Fig. 3.9(a) due to diffuse scattering. Compared to Fig. 3.6(a) and (b) the facets have a much higher height, which explains why there is no intensity reduction observed in the x-ray reflectometry measurements of GGG/YIG samples (cf. Fig. 3.4(a)). More details about the formation of facets are discussed in Sec. 3.7.

On the other hand Pt thin films annealed at $350^\circ$C show no facets and no difference in the surface roughness (cf. Fig. 3.10(d)) compared to the surface topography of a Pt thin film without annealing (see Fig. 3.10(a)). Furthermore, the determined roughnesses match to the one in Fig. 3.9(b) since a flat surface yields the same results for a line profile or a topography map.

### 3.3.3 Conclusion

Once again a substrate temperature of $350^\circ$C reveals the best results for the fabrication of a trilayer focusing on the Pt behavior. First this temperature is the same as in Sec. 3.2.5 leading to optimal results for a YIG thin film grown in argon on GGG. Secondly the Pt thickness fringes are still visible in the reflectometry in Fig. 3.9(a), which is important for a reliable simulation to determine the actual deposited Pt thickness. And thirdly no facets form as shown by the AFM.
3.4 $\text{Y}_3\text{Fe}_5\text{O}_{12}$ on Pt thin films on $\text{Gd}_3\text{Ga}_5\text{O}_{12}$

The next step in the fabrication of YIG|Pt|YIG trilayers is to investigate YIG thin films grown on Pt layers (cf. inverse bilayer in Fig. 3.1(d)). This is very interesting because here a crystalline YIG thin film is tried to grow on a polycrystalline Pt layer. Thus, a crystalline substrate is no longer existent. The fabrication starts in the same way as described in Sec. 3.3 with the deposition of a Pt thin film on GGG using EVAP. After the sample is transferred into the PLD chamber a crystalline YIG thin film is tried to be grown onto the Pt layer using an argon at-
mosphere of 25 µbar pressure and substrate temperatures of 550 °C, 450 °C or 350 °C. The next section describes the structural properties of the fabricated GGG|Pt|YIG inverse bilayers using x-ray diffractometry.

### 3.4.1 X-ray diffractometry

#### 2θ − ω scans, out-of-plane lattice constants, reflectometries and rocking curves

The measured 2θ − ω scans of the YIG thin film on Pt layers are shown in Fig. 3.11(a). Since no YIG (444) reflection is visible for a substrate temperature of 350 °C, an additional sample is fabricated using an oxygen atmosphere instead of argon (cf. blue curve in Fig. 3.11(a)). But the blue curve shows no change, which indicates that missing oxygen is not the reason. For all four samples a Pt (111) reflection is visible in the 2θ − ω scan and behaves in the same way as described in Sec. 3.3.1. The only difference is a higher intensity for Ts ≤ 450 °C. The YIG (444) reflexes show a clear trend in Fig. 3.11(a): for a decreasing substrate temperature the peak intensity increases.

![Figure 3.11](image)

**Figure 3.11:** (a) 2θ − ω scans of the GGG|Pt|YIG inverse bilayers. Since no YIG (444) reflection is visible for a substrate temperature of 350 °C an additional sample is fabricated in an oxygen atmosphere instead of argon (with the same pressure). For all these different samples the out-of-plane lattice constants a are calculated as described in Sec. 3.2.2 for the four Pt (111) reflections and the two YIG (444) reflections. The received values are displayed in the lower (aPt) and upper (aYIG) plot part of (b) versus the substrate temperature Ts. (c) Measured x-ray reflectometries. (d) Rocking curves of the marked YIG (444) reflexes in (a) which are shifted (Δω) as described in Fig. 3.3. Additionally Gaussian fits are sketched to determine the full width at half maximums (FWHM).
intensity reduces and vanishes at 350 °C. At this temperature YIG does not grow in a detectable crystalline form on the Pt layer independent of the used gas atmosphere. Furthermore, the visible YIG reflections show no Laue oscillations, which indicates no coherent growth of the lattice planes. The crystalline quality of YIG thin films grown on polycrystalline Pt layers is worse compared to YIG thin films grown on GGG substrates (cf. Fig. 3.2(b)).

Figure 3.11(b) shows the calculated out-of-plane lattice constants from the Pt (111) and the YIG (444) reflexes in Fig. 3.11(a) as described in Sec. 3.2.2. The received Pt lattice constants of the GGGPtYIG samples fabricated at 350 °C match well with the literature value [31] and slightly differ for inverse bilayers fabricated at higher substrate temperatures. For the YIG lattice constants also oxygen vacancies as detected in the GGGYIG samples are responsible for higher values compared to the literature value of bulk YIG [27, 28]. Furthermore, no trend depending on the substrate temperature can be detected since only two values are available.

The measured reflectometries for all four samples are shown in Fig. 3.11(c). As expected from Sec. 3.3 no Pt thickness fringes are visible for samples with $T_S = 550 °C$ and only the ones from YIG remain. This changes for a substrate temperature of 450 °C where in contrast to Fig. 3.9(a) Pt oscillations are still apparent and a higher intensity for larger angels occurs. A common property of both substrate temperatures is the missing loss of intensity for small $2\theta$ angles. On the other hand, Pt thickness fringes are visible as expected for a substrate temperature of 350 °C independent of the used gas atmosphere during growth. The differences between both samples are better distinctive YIG oscillations and wider Pt thickness fringes in the blue curve. The first one could be caused by a more coherent growth of YIG in an oxygen atmosphere. The existence of YIG oscillations in the reflectometry but no reflex in the $2\theta - \omega$ scan shows that non-crystalline YIG grows on Pt for a substrate temperature of 350 °C independent of the used gas atmosphere during growth.

The rocking curves corresponding to the two marked YIG (444) reflections in Fig. 3.11(a) are shown in Fig. 3.11(d). For a substrate temperature of $T_S = 550 °C$ the rocking curve consists of two components. The first one reveals a low mosaic spread of the YIG lattice planes while the second one yields a high mosaic spread, which is probably caused by the first YIG lattice planes growing on the Pt layer. In case of the GGGPtYIG sample fabricated at 450 °C the rocking curve around the YIG (444) reflection reveals only one component due to the very low intensity. The FWHM values for the component of a low mosaic spread of the inverse bilayers are larger than the corresponding values for the GGGYIG samples (cf. Fig. 3.3(b)), which indicates a reduced crystalline quality of the YIG thin films on polycrystalline Pt.

Simulations

The simulations of the reflectometries of the samples fabricated with a substrate temperature of 350 °C either in oxygen or argon are shown in Fig. 3.12. No good agreement between experiment and simulation is obtained by using the ideal system GGGPtYIG (cf. red dashed line in Fig. 3.12(a)) with a Pt thickness of 7 nm. While the YIG oscillations can be well fitted for $2\theta < 1°$ this is not possible for the Pt oscillations even for varying the thickness over a wide range. Decreasing the density of Pt ($\rho_{Pt} = 21.4$ g cm$^{-3}$ [31]) in the ideal simulation towards the value of YIG ($\rho_{YIG} = 5.17$ g cm$^{-3}$ [28]) improves the fit quality of the Pt oscillations. Therefore, platinum oxide (PtO) with a density of $\rho_{PtO} = 14.9$ g cm$^{-3}$ [40] is used in another simulation to check whether the Pt layer oxidizes during the deposition of YIG. The system GGGPtOYIG (cf. blue dashed line in Fig. 3.12(a)) is able to fit the first Pt oscillation but differs for larger $2\theta$ angles using a thickness of about 11 nm for PtO. First the combination of Pt and PtO in the simulation GGGPtOYIG (cf. black dashed line in Fig. 3.12(a)) is able to fit all Pt oscillations but does not agree with the height of them and, thus, the roughness for $2\theta > 2°$. This simulation uses a Pt thickness of 0.8 nm and 8.7 nm for PtO.
Figure 3.12: (a) and (b) Simulations of the two reflectometries with a substrate temperature of 350°C from Fig. 3.11(c). Each plot contains three different simulations: the first one uses the ideal system GGG|Pt|YIG (red dashed line) with a maximal thickness for Pt of about 7 nm as determined in Fig. 3.9(b). The second one uses a layer of platinum oxide (PtO) between GGG and YIG (blue dashed line in (a) and olive dashed line in (b)) and the third simulation combines Pt and PtO between the substrate GGG and the YIG thin film (black dashed line).

Anyway the assumption of PtO is just a try to improve the fit curves but any other chemical system is possible for the interface area between Pt and YIG or even between GGG and Pt (e.g. diffusions or reactions). Here additional experiments as resistance measurements or element specific investigations are mandatory to get a better insight. A definite statement following from the simulations in Fig. 3.12(a) is that the ideal GGG|Pt|YIG inverse bilayer does not exist in the sample fabricated in argon at $T_S = 350°C$. The same results apply for the YIG thin films on Pt layers using substrate temperatures of 550°C and 450°C where it is even more difficult to simulate the reflectometries. Here only reliable values for the YIG thicknesses $d_{YIG}$ can be extracted which are shown in Tab. 3.1 with the comparison to those of the YIG thin films grown directly on GGG from Sec. 3.2.2.

| gas atmosphere | substrate temperature $T_S$ | $d_{YIG}$ for GGG|YIG | $d_{YIG}$ for GGG|Pt|YIG |
|----------------|-----------------------------|---------------------|---------------------|
| Ar             | 550 °C                      | (49 ± 1) nm         | (62.5 ± 1.0) nm     |
| Ar             | 450 °C                      | (55.6 ± 0.4) nm     | (42.9 ± 1.0) nm     |
| Ar             | 350 °C                      | (50.4 ± 0.4) nm     | (34.8 ± 0.5) nm     |
| O$_2$          | 550 °C                      | (41.8 ± 0.2) nm     | –                   |
| O$_2$          | 350 °C                      | –                   | (44.3 ± 0.2) nm     |

Table 3.1: YIG thickness comparison between GGG|YIG and GGG|Pt|YIG samples.

Table 3.1 reveals that the YIG growth in argon is worsen for a decreasing substrate temperature $T_S$ on polycrystalline Pt due to the missing crystalline substrate GGG offering a small lattice mismatch along the (111) direction. On the other hand, the thicknesses of the YIG films grown in an oxygen atmosphere seem to be independent of the substrate temperature as well as of a non-crystalline layer underneath.

The different simulation steps for the inverse bilayer grown in oxygen (cf. Fig. 3.12(b)) are identical with the above discussed steps for Fig. 3.12(a) and show the same result using the ideal system GGG|Pt|YIG. An essential difference is the match between the GGG|PtO|YIG ($d_{PtO} \approx 7.6$ nm) and GGG|Pt|PtO|YIG ($d_{Pt} \approx 0.7$ nm and $d_{PtO} \approx 6.6$ nm) simulation with the
reflectometry. This indicates that Pt oxidizes in an oxygen atmosphere and, thus, that the assumption of PtO is appropriate. Furthermore, the choice of an argon atmosphere, as mentioned in Sec. 3.2.1 to avoid or at least minimize the oxidation of Pt, is justified. The next section describes the surface topographies of the inverse bilayers using atomic force microscopy.

### 3.4.2 Atomic force microscopy

The measured surface topographies of the different GGG|Pt|YIG inverse bilayers are shown in Fig. 3.13. As in Fig. 3.6 and 3.10 facets are visible in Fig. 3.13(a) and (b) for a substrate temperature equal or greater than 450 °C and are discussed in more detail in Sec. 3.7.

![Figure 3.13: (a), (b) and (c) Surface topographies for the GGG|Pt|YIG inverse bilayers grown in argon at three different substrate temperatures and (d) for the inverse bilayer grown in oxygen. In (a) and (b) once again facets are visible. These are missing in (c) and (d) leading to a flat surface. The rougher part in the upper middle in (d) is probably caused by dirt while the peak in (c) with a height of about 35 nm is raised by a too high feedback signal or a sticked tip during the AFM measurement.](image)

Figure 3.13(c) and (d) do not show these facets. The determined roughness in Fig. 3.13(c) matches with the one of the simulation in Fig. 3.12(a). Furthermore, this value is higher com-
pared to the surface roughness of the YIG thin film on GGG in Fig. 3.6(c). The next section describes the magnetic properties of the GGG|Pt|YIG samples using SQUID magnetometry.

### 3.4.3 SQUID magnetometry

The measured magnetic hysteresis curves for all four inverse bilayers are shown in Fig. 3.14(a) and the extracted saturation magnetizations and coercive fields in Fig. 3.14(b) (as described in Sec. 3.2.4). These values clearly differ from those for YIG thin films grown on GGG substrates (cf. Fig. 3.7(b)). The saturation magnetization of a YIG thin film grown on Pt at a substrate temperature of $T_S = 550°C$ is already slightly smaller compared to a YIG thin film fabricated with the same conditions on a GGG substrate. This difference is more pronounced at $T_S = 450°C$. At $T_S = 350°C$ no ferrimagnetic ordering of YIG is detectable independent of the gas atmosphere. The fact that also the inverse bilayer fabricated in oxygen has no ferrimagnetic ordering shows that this phenomena is not caused by the gas atmosphere analogous to its influence on the crystalline growth in the $2θ − ω$ scan in Fig. 3.11(a). Furthermore, the calculated saturation magnetizations are much smaller compared to the literature value [25]. This difference is caused by the worse crystalline quality of the YIG thin films. Also the coercive fields of the inverse bilayers fabricated with a substrate temperature of 550°C and 450°C increase by a factor of about two indicating a higher density of defects in the YIG thin films on Pt.

### 3.4.4 Conclusion

So far a substrate temperature of 350°C is most convenient for the fabrication of a trilayer according to Sec. 3.2.5 and Sec. 3.3.3. But YIG thin films grown at 350°C on Pt layers do not show any ferrimagnetic ordering and are not crystalline anymore. Thus, this temperature is not suitable to fabricate a trilayer, which can be used for example as spin valve where two adjustable magnetization in both YIG thin films are required (see Sec. 2.1).
3.5 Pt on $Y_3Fe_5O_{12}$ thin films on Gd$_3$Ga$_5$O$_{12}$

The last fabrication step in Fig. 3.1(e) deals with the deposition of Pt on a YIG thin film that is grown on GGG in the PLD chamber. These systems are already well examined in Ref. [10] showing that the deposition of Pt in the EVAP has no influence on the physical properties of YIG. Changes might occur when the substrate is reheated in the last fabrication step of a trilayer to grow the second YIG layer. This is investigated in the next section.

3.6 Trilayer on Gd$_3$Ga$_5$O$_{12}$

Although the YIG thin film grown on Pt at a substrate temperature of 350 °C does not show a ferrimagnetic ordering as mentioned in Sec. 3.4.4, a trilayer is fabricated using this temperature for the growth of both YIG layers in an argon atmosphere. The fabrication starts as described in Sec. 3.5 followed by an additional PLD process for the second YIG layer (cf. YIG2 in Fig. 3.1(a)). Thus, the so far collected properties of the different sample types in Fig. 3.1(b)–(e) can be reviewed for the structural, surface and magnetic behavior of the trilayer.

3.6.1 X-ray diffractometry

$2\theta - \omega$ scan and rocking curves

The $2\theta - \omega$ scan of the trilayer is shown in Fig. 3.15(a). The Pt (111) reflection behaves like the one described in Sec. 3.3.1 for an annealing temperature of 350 °C. The YIG (444) peak shows a high crystalline quality proven by the symmetric Laue oscillations and belongs two the first YIG layer since the second one does not grow crystalline (cf. Fig. 3.11(a)). Calculating the out-of-plane lattice constants as described in Sec. 3.2.2 gives $a_{YIG} = (1.2452 \pm 0.0001)$ nm and $a_{Pt} = (0.3883 \pm 0.0005)$ nm. Both values differ slightly from the literature ones [27, 28, 31]. The difference for YIG is caused by oxygen vacancies in the unit cell.

Figure 3.15(b) shows the corresponding rocking curves belonging to the marked reflexes in Fig. 3.15(a). The Pt (111) reflection behaves like

![Figure 3.15: (a) $2\theta - \omega$ scan of the trilayer. (b) Corresponding rocking curves for the Pt (111) and YIG (444) reflection which are shifted ($\Delta \omega$) as described in Fig. 3.3(a) for a better comparison.](image)
Reflectometry and Simulation

The measured reflectometry and the corresponding simulation of the trilayer are both shown in Fig. 3.16. As expected from Fig. 3.9(a) and Fig. 3.11(c) Pt thickness fringes (larger ones) are visible which are superimposed by YIG thickness oscillations caused from both layers. As described in Sec. 3.4.1 three different heterostructures are used trying to simulate the reflectometry. The additional layer PtO is assumed between Pt and the upper YIG thin film YIG2. The ideal system GGG|YIG1|Pt|YIG2 is not able to describe the trilayer as does the simulation GGG|YIG1|PtO|YIG2 assuming pure PtO since the roughness of the YIG oscillations for larger angles is not correct. Here the combination of Pt and PtO leads to an excellent fit (GGG|YIG1|Pt|PtO|YIG2). But as described in Sec. 3.4.1 further investigations (especially a resistance measurement of the Pt layer) of the occurring interfaces YIG1|Pt and Pt|YIG2 are necessary to gain a better insight. The next section describes the surface and magnetic properties of the fabricated trilayer.

3.6.2 Atomic force microscopy and SQUID magnetometry

The surface topography and the measured magnetic hysteresis of the trilayer are both shown in Fig. 3.17. The roughness in Fig. 3.17(a) confirms a flat but rough surface of the trilayer as expected by the AFM measurement of a single YIG thin film (cf. Fig. 3.6(c)) an annealed Pt thin film (cf. Fig. 3.10(d)) and an inverse bilayer (cf. Fig. 3.13(c)) fabricated at $T_S = 350°C$.

As described in Sec. 3.2.4 the saturation magnetization and coercive fields are calculated from the measured magnetic hysteresis in Fig. 3.17(b). Since the second YIG layer of the trilayer has no magnetic ordering according to Fig. 3.14(b) the first one is responsible for the measured curve. This is confirmed by Fig. 3.7(b) since the saturation magnetizations match within their error as do the coercive fields.

3.6.3 Conclusion

The previous discussions show that the so far developed system from Sec. 3.2, 3.3 and 3.4 can describe the experimental results of the trilayer grown on GGG. Anyway this means that nothing
3.7 Formation of facets of thin films on Gd$_3$Ga$_5$O$_{12}$

For substrate temperatures equal or greater than 450 °C facets have formed for GGG|YIG in Fig. 3.6, GGG|Pt in Fig. 3.10 and the inverse bilayer GGG|Pt|YIG in Fig. 3.13. These facets could be explained by a three dimensional growth of YIG (Volmer-Weber growth [41]) due to a higher energy (sum of surface and interface energy) compared to the substrate surface en-

Figure 3.17: (a) Surface topography and (b) measured magnetic hysteresis of the trilayer. Only the volume of the first YIG layer (YIG1) is used to calculate the magnetization $M$ over Eq. (A.3) since the second one is expected to not show any magnetic ordering according to Fig. 3.14(b).

has changed about the missing ferrimagnetic ordering of the second YIG layer. Therefore, another substrate is tested in the next chapter 4. But before the next section discusses the formation of facets of thin films deposited on the substrate GGG.

3.7 Formation of facets of thin films on Gd$_3$Ga$_5$O$_{12}$

For substrate temperatures equal or greater than 450 °C facets have formed for GGG|YIG in Fig. 3.6, GGG|Pt in Fig. 3.10 and the inverse bilayer GGG|Pt|YIG in Fig. 3.13. These facets could be explained by a three dimensional growth of YIG (Volmer-Weber growth [41]) due to a higher energy (sum of surface and interface energy) compared to the substrate surface en-

Figure 3.18: (a) and (b) Comparison of the surface topographies of the substrate GGG before and after the annealing for one hour at a temperature of 550 °C using an argon atmosphere of 25 μbar pressure.
nergy. This growth mode could be examined by using reflection high-energy electron diffraction (RHEED) [38, 39] during the YIG deposition. Furthermore, this assumed behavior of YIG is in agreement with the one of bismuth iron garnet Bi$_3$Fe$_5$O$_{12}$ (BIG) grown on (111)-orientated GGG substrates as described in Ref. [42]. This article assumes that growing BIG trends to form (110) facets. However, since facets form for YIG and Pt they could be caused by the substrate GGG itself. As soon as a thin film is coating the substrate surface a temperature equal or greater than 450°C causes a surface reconstruction of GGG. Thereby the garnet tries to minimize its free energy by forming facets into the (110) direction being the preferred one. This could explain the detected behavior in Fig. 3.6, 3.10 and 3.13 without disagreeing to the result of an annealing process of a GGG substrate for one hour at $T_S = 550°C$ in argon (cf. Fig. 3.18).

Therefore, the next chapter uses another substrate trying to minimize the formation of facets or at least to reach a higher substrate temperature $T_S$ where facets start to form for YIG and Pt thin films. Thus, the YIG thin films could be grown at higher substrate temperatures, which might improve the magnetic properties. Especially for the second YIG layer of the trilayer a better ferrimagnetic ordering could be reached in this way.
Chapter 4

\( Y_3Fe_5O_{12}|Pt|Y_3Fe_5O_{12} \) trilayer on \( Y_3Al_5O_{12} \) substrate

Since no uniform growth temperature for both YIG layers has been found on a GGG substrate to achieve the described properties for the trilayer in Sec. 2.1, YAG is used as another substrate. For the growth of YIG in an oxygen atmosphere on (111)-orientated YAG substrates a temperature of 500 °C is found to be the optimized one according to Ref. [10]. Thus, the same optimization steps as described in chapter 3 are performed to find the optimal substrate temperature \( T_S \) for an argon atmosphere with 25 µbar pressure to avoid the oxidation of Pt during the last fabrication step of the trilayer. The next section describes the behavior of YAG as substrate material towards heat compared to the substrate GGG.

4.1 \( Y_3Al_5O_{12} \) as substrate material

The AFM measurements of the substrate YAG before and after the annealing for one hour at \( T_S = 550 \) °C in an argon atmosphere with 25 µbar pressure are shown in Fig. 4.1.

![Figure 4.1: Comparison of the surface topographies of the substrate YAG before and after the annealing for one hour at a temperature of 550 °C in an argon atmosphere. The oscillating lines in both pictures are caused by the cantilever tip sticking to the substrate as described in Sec. A.2.2.](image)

While GGG seems to get rougher (cf. Fig. 3.18(b)), YAG reduces its surface roughness as it is expected by an annealing process (cf. Fig. 4.1(b)). Thus, the surface energy of YAG and GGG should be different altering the substrate temperature when facets start to form. This
result is decisive for the choose of YAG as the next substrate to fabricate a YIG|Pt|YIG trilayer. Therefore, the next section discusses the growth examination of YIG thin films (cf. Fig. 3.1(b)) using the substrate YAG instead of GGG.

4.2  \( \text{Y}_3\text{Fe}_5\text{O}_{12} \) thin films on \( \text{Y}_3\text{Al}_5\text{O}_{12} \)

Analogous to Sec. 3.2 three different substrates temperatures of \( T_S = 550 \, ^\circ\text{C}, 450 \, ^\circ\text{C} \) and \( 350 \, ^\circ\text{C} \) are used to fabricate YIG thin films in argon on the substrate YAG. Furthermore, an additional reference sample is fabricated with the optimal parameters for an oxygen atmosphere from Ref. [10] for comparison. The following sections describe the structural, surface and magnetic properties of the YIG thin films and compare the results to the YIG on GGG series.

4.2.1  X-ray diffractometry

\( 2\theta – \omega \) scans and out-of-plane lattice constants

The \( 2\theta – \omega \) scans for the temperature series in argon and the reference sample grown in oxygen are shown in Fig. 4.2(a). Due to the higher lattice mismatch between YAG and YIG compared to GGG and YIG (see Sec. 2.2) the YIG (444) reflexes have a larger distance from the substrate reflection YAG (444). Furthermore, the intensity of the YIG reflexes is much lower compared to those in Fig. 3.2(b), which is also caused by the higher lattice mismatch and, thus, a worse crystalline quality of the YIG thin films. A reduced peak intensity is also visible compared to the reference sample, which indicates the influence of the gas atmosphere on the crystalline quality. In addition, Laue oscillations are missing in the \( 2\theta – \omega \) scans compared to the YIG thin films on GGG (cf. Fig. 3.2(b)). For a substrate temperature of \( 350 \, ^\circ\text{C} \) no peak is visible in Fig. 4.2(a) indicating no crystalline growth of YIG on YAG at this temperature.

The out-of-plane lattice constants are calculated as described in Sec. 3.2.2 from the marked YIG (444) reflexes in (a).
4.2 \( Y_3Fe_5O_{12} \) thin films on \( Y_3Al_5O_{12} \)

### Rocking curves and full width at half maximums

The measured rocking curves at the marked YIG (444) reflexes in Fig. 4.2(a) are shown in Fig. 4.3(a). The extracted full width at half maximums (FWHM) in Fig. 4.3(b) of the two components in the rocking curves are comparable with those from GGG|YIG in Fig. 3.3(b). The low mosaic spread components are also smaller than 0.04° indicating a high crystalline component of the corresponding lattice planes. However, the intensity of the higher mosaic spread components is larger than for YIG thin films grown on GGG revealing tilted lattice planes, which could be caused by the high lattice mismatch for the first growing YIG planes.

![Figure 4.3:](image)

**Figure 4.3:** (a) Rocking curves around the marked YIG (444) reflexes in Fig. 4.2(a). The rocking curves are shifted (\( \Delta \omega \)) as described in Fig. 3.3(a). (b) Full width at half maximums (FWHM) extracted from the two components visible in each rocking curves. All FWHM values are plotted versus the corresponding substrate temperature \( T_S \).

### Reflectomtries and simulations

The measured reflectometries are shown in Fig. 4.4(a). For all temperatures except 350°C YIG thickness oscillations are visible. For a substrate temperature of 350°C different chemical compounds (one or more) than YIG seem to grow on the substrate YAG causing the different reflectometry. For the other curves the corresponding simulations are shown in Fig. 4.4(b), (c) and (d), which excellently agree with the experimental data. The extracted YIG thicknesses reveal a trend depending on the used gas atmosphere during the growth. While the reference sample has a thickness comparable to those for YIG thin films on GGG (cf. Fig. 3.5), the two YIG thin films grown in argon have a much smaller one. Thus, the different gas atmosphere has an essential impact on the growth quality of YIG thin films on YAG.

On the other hand, the extracted surface roughnesses seem to match for all three simulations and are consequently independent of the used gas atmosphere. But this result is examined in more detail in the next section using atomic force microscopy.

### 4.2.2 Atomic force microscopy

The measured surface topographies of the YIG thin films are shown in Fig. 4.5. Figure 4.5(a) reveals that also on YAG facets form but not until a substrate temperature of 550°C. Therefore, it seems that the surface energy of YIG is different from that of GGG making YAG substrates more suitable for the fabrication of YIG|Pt|YIG trilayers.
Figure 4.4: (a) Measured x-ray reflectometries for YIG thin films grown in argon and the reference sample grown in oxygen on YAG. The corresponding simulations are shown in (b)–(d) with the extracted YIG thickness $d_{\text{YIG}}$ and surface roughness $R_{\text{RMS}}$.

For a substrate temperature of 350 °C a very rough surface with holes of about 30 nm is visible in Fig. 4.5(c). This result matches to the corresponding reflectometry in Fig. 4.4(a) showing that probably a thin film with a different stoichiometry than YIG has grown on YAG.

Figure 4.5(b) and (d) reveal that YIG can grow on YAG with a smaller surface roughness compared to YIG thin films on GGG in Fig. 3.6, without forming any facets and independent of the used gas atmosphere (argon or oxygen). Based on this result the magnetic hysteresis is only measured for a substrate temperature of 450 °C in the next section using SQUID magnetometry.

### 4.2.3 SQUID magnetometry

The measured magnetic hysteresis for the YIG thin film grown in argon at a substrate temperature of 450 °C is shown in Fig. 4.6. Since YAG is diamagnetic, the splitting of the data points in the area of the saturation magnetization $M_S$ is missing compared to Fig. 3.7(a) after the subtraction of the substrate background as described in Sec. 3.2.4. Consequently the mentioned error in Eq. (3.3) can not be neglected here and is additionally added to $M_S$.

The calculated saturation magnetization $M_S$ matches within its uncertainty to the one from Fig. 3.7(b) for a substrate temperature of 450 °C. The same applies for the coercive field $\mu_0H_C$. Thus, the worse crystalline quality of YIG thin films on YAG in argon has no influence on the magnetic properties compared to the reference sample or to YIG thin films grown on GGG.
4.2 \( \text{Y}_3\text{Fe}_5\text{O}_{12} \) thin films on \( \text{Y}_3\text{Al}_5\text{O}_{12} \)

**Figure 4.5:** (a)–(d) Surface topographies of YIG thin films grown on YAG.

**Figure 4.6:** Magnetic hysteresis measured at 300 K of a YIG thin film grown on YAG at 450°C in argon (red symbols) as well as a YIG thin film fabricated at 500°C in oxygen (blue symbols). The saturation magnetization \( M_S \) of YIG is calculated as described in Sec. 3.2.4 using the data points with \(|\mu_0 H_C| \geq 3\text{T} \). This yields \( M_S = (115.5 \pm 2.8) \text{kA m}^{-1} \) and \( M_S = (100.7 \pm 3.4) \text{kA m}^{-1} \) for the YIG thin film grown in argon and oxygen, respectively. The coercive fields are \( \mu_0 H_C = (2.9 \pm 0.1) \text{mT} \) and \( \mu_0 H_C = (4.1 \pm 0.1) \text{mT} \) for each sample. The reference sample grown in oxygen at 500°C is taken from Ref. [43] with the sample id “YY15”. 

\[ \mu_0 H_C = (2.9 \pm 0.1) \text{mT} \text{ and } \mu_0 H_C = (4.1 \pm 0.1) \text{mT} \]
4.2.4 Conclusion

For the growth of YIG thin films on YAG a substrate temperature of 450 °C is most convenient. The reasons are a small surface roughness, no forming facets (cf. Fig. 4.5(b)) and magnetic properties which are comparable to those from YIG on GGG although a reduced crystalline growth quality exists (cf. Fig. 4.2(a) and 4.6). Since for a substrate temperature of 350 °C a thin film with a different stoichiometry than YIG grows on YAG, this specific temperature is no longer examined in the following sections.

4.3 Pt thin films on Y₃Al₅O₁₂

This fabrication step (cf. Fig. 3.1(c)) has the same purpose as described in Sec. 3.3 only using YAG instead of GGG and annealing temperatures of 550 °C and 450 °C. The next section describes the structural properties of the annealed Pt thin films using x-ray diffractometry.

4.3.1 X-ray diffractometry

$2\theta - \omega$ scans and rocking curves

The measured $2\theta - \omega$ scans of Pt thin films annealed at 550 °C and 450 °C for one hour in argon are shown in Fig. 4.7(a). A similar behavior of the Pt (111) reflection as described in Sec. 3.3.1 is visible. For an annealing temperature of 550 °C a very high intensity component is visible indicating the growth of large crystallites. Calculating the out-of-plane lattice constant according to Sec. 3.2.2 gives $a_{Pt} = (0.3926 \pm 0.0050)$ nm and $a_{Pt} = (0.3922 \pm 0.0005)$ nm for the Pt thin film annealed at 450 °C and 550 °C, respectively. Both values perfectly agree with the literature value of 0.3920 nm [31].

The rocking curves in Fig. 4.7(b) show that for an annealing temperature of 450 °C Pt is still polycrystalline but it also exists a narrow component with a very low mosaic spread indicated by a FWHM smaller than 0.04 °. The corresponding crystallites have a high crystalline quality, which causes the higher intensity in the $2\theta - \omega$ scan compared to Pt thin films on GGG annealed at the same temperatures (cf. Fig. 3.8(a)). For an annealing temperature of 550 °C the much larger grown crystallites cause a higher intensity of the corresponding rocking curve. The large
4.3 Pt thin films on \( Y_3Al_5O_{12} \)

FWHM of 0.41° shows that the lattice planes have a high mosaic spread but also the approach of a small mosaic spread component is visible around \( \Delta \omega = 0 \).

Compared to Fig. 3.8(b) the crystalline quality of the annealed polycrystalline Pt thin films is improved on YAG than on GGG due to the lower lattice mismatch as calculated in Sec. 2.2.

Reflectometries and simulations

The measured reflectometries for both samples are shown in Fig. 4.8(a). This result is similar to annealed Pt thin films on GGG (cf. Fig. 3.9(a)). For an annealing temperature of 450 °C the Pt thickness oscillations are still visible while they vanish for an annealing temperature of 550 °C and an additional loss of intensity occurs. Possible reasons have already been discussed in Sec. 3.3.1.

Figure 4.8(b) shows the simulation of the reflectometry corresponding to an annealing temperature of 450 °C. The extracted Pt thickness is the same as in Fig. 3.9(b) for an annealing temperature of 350 °C and the determined surface roughness is confirmed in the next section using atomic force microscopy.

**4.3.2 Atomic force microscopy**

The measured surface topographies of the annealed Pt thin films are shown in Fig. 4.9. As detected in Fig. 3.10(b) for the annealing of Pt on GGG, facets are also visible for an annealing temperature of 550 °C in Fig. 4.9(a) for Pt on YAG. These facets probably cause the intensity loss of the reflectometry in Fig. 4.8(a). For an annealing temperature of 450 °C no facets are visible and the determined surface roughness matches to the one in Fig. 4.8(b).

**4.3.3 Conclusion**

The annealing of Pt thin films on YAG substrates manifests one specific temperature that is suitable for the fabrication of a trilayer on YAG. According to the reflectometries in Fig. 4.8 and the AFM measurements in Fig. 4.9 only a substrate temperature of 450 °C can be used for the growth of the second YIG layer. Interestingly this temperature is higher than for the optimal temperature of annealed Pt thin films on GGG discussed in Sec. 3.3. Furthermore, it matches the optimized one in Sec. 4.2.4. Therefore, the following fabrication step of YIG thin
4.4 \( \text{Y}_3\text{Fe}_5\text{O}_{12} \) on Pt thin films on \( \text{Y}_3\text{Al}_5\text{O}_{12} \)

The YIG thin film on a Pt layer is fabricated at a substrate temperature of 450 °C in argon as described in Sec. 3.4. This is the decisive step towards the fabrication of trilayers whether a ferrimagnetic ordering of the second YIG thin film is measurable using the substrate YAG instead of GGG. The next section describes the structural properties of the fabricated sample.

4.4.1 X-ray diffractometry

\( 2\theta - \omega \) scan and rocking curve

The measured \( 2\theta - \omega \) scan for the YIG thin film on a Pt layer grown on YAG at a substrate temperature of 450 °C is shown in Fig. 4.10(a). The Pt (111) reflection develops as described in Sec. 3.3.1. The rocking curve in Fig. 4.10(b) reveals that Pt is still polycrystalline and compared to Fig. 4.7(b) the low mosaic spread component is missing. Calculating the out-of-plane lattice constant of the Pt (111) reflex as described in Sec. 3.2.2 yields \( a_{\text{Pt}} = (0.3881 \pm 0.0090) \text{ nm} \) which matches within the uncertainty to the literature value [31]. Furthermore, no YIG (444) reflection is visible in Fig. 4.10(a) indicating no crystalline growth of YIG on polycrystalline Pt deposited on the substrate YAG. This result is comparable to GGG|Pt|YIG inverse bilayer in Fig. 3.11(a) for a substrate temperature of 350 °C.

Reflectometry and simulation

The measured x-ray reflectometry with the corresponding simulation is shown in Fig. 4.11. As expected Pt thickness fringes are visible superimposed by YIG oscillations in Fig. 4.11(a). Analogous to Sec. 3.4.1 three different simulations are used in Fig. 4.11(b) trying to fit the experimental data. The ideal heterostructure YAG|Pt|YIG is not able to describe the measured
Figure 4.10: (a) \(2\theta - \omega\) scan of the YAG|Pt|YIG sample. (b) Measured rocking curve around the marked Pt (111) reflection.

Figure 4.11: (a) X-ray reflectometry of the YIG thin film on Pt layer on YAG and (b) corresponding simulations. For the ideal system (dashed green line) the maximal possible Pt thickness of 7 nm is used according to Fig. 3.9(b). The second simulation (blue dashed line) assumes PtO with a thickness of about 9.3 nm. The combination of Pt and PtO in the third simulation (black dashed line) uses a thickness of 0.4 nm for Pt and 8.8 nm for PtO.

reflectometry. Assuming pure PtO instead of Pt between YAG and YIG shows a great improvement of the simulation but differs for larger \(2\theta\) angles. The combination of Pt and PtO results in a simulation which excellently matches. Since the simulation of the reflectometry is sensitive to the density of the assumed material system, the layer in between Pt and YIG could also be an intermixing of Pt and YIG creating a Pt doped YIG. Also here further experiments are necessary to identify the interface Pt|YIG.

Comparing the extracted YIG thickness of \((22.3 \pm 0.2)\) nm to the one for YIG thin films on YAG (cf. Fig. 4.4(c)) shows that the YIG growth on polycrystalline Pt is worse. Furthermore, the surface roughness of \((1.5 \pm 0.1)\) nm seems to increase but this is measured in more detail in the next section using atomic force microscopy.

4.4.2 Atomic force microscopy and SQUID magnetometry

The measured surface topography of the inverse bilayer using atomic force microscopy is shown in Fig. 4.12(a). No facets are visible but the surface is rougher compared to YIG thin films on
YAG on Pt on YIG. (b) Magnetic hysteresis with the extracted saturation magnetization and coercive field as described in Sec. 3.2.4.

YAG (cf. Fig. 4.5(b)) and slightly differs from the value determined by simulating the x-ray reflectometry in Fig. 4.11(b).

The result of the measurement of the magnetic hysteresis of this sample is the decisive step towards the fabrication of trilayers to achieve the described properties in Sec. 1. A YIG thin film grown on polycrystalline Pt has shown no ferrimagnetic ordering in case of GGG|Pt|YIG inverse bilayers for a substrate temperature of 350 °C as discussed in Sec. 3.4.3. Therefore, it is interesting whether the change of the substrate from GGG to YAG causes a different result.

Figure 4.12(b) clearly shows that for the so far convenient substrate temperature of 450 °C a ferrimagnetic ordering of YIG grown on polycrystalline Pt is measurable. Compared to YAG|YIG in Fig. 4.6 the saturation magnetization $M_S$ is only slightly lower. Furthermore, $M_S$ is much larger than for all substrate temperatures in Fig. 3.14 for the inverse bilayers on GGG. A clear difference to YIG on YAG is a much larger coercive field indicating a harder magnetic YIG thin film. In addition, the measurable hysteresis shows that a non crystalline growth of YIG on Pt is not the main reason for the missing ferrimagnetic ordering in Fig. 3.14(a).

### 4.4.3 Conclusion

Using a substrate temperature of 450 °C shows that in contrast to the GGG series a consistent growth temperature exists where both YIG thin films in a trilayer structure have a measurable ferrimagnetic ordering. The next section describes the annealing of a Pt layer deposited on a YIG thin film on YAG to examine the influence of the reheating process onto the interface between the YIG and Pt layer.

### 4.5 Pt on $Y_3Fe_5O_{12}$ thin films on $Y_3Al_5O_{12}$

As described in Sec. 3.5 the deposition of Pt in the EVAP on a YIG thin film has no influence on the physical properties of YIG. This might change when the substrate is reheated again. This situation is examined in the following sections. Therefore, a YIG thin film is grown in argon at a substrate temperature of 450 °C and a Pt film with an set thickness of 10 nm is deposited onto it in the EVAP to achieve the sample type shown in Fig. 3.1(e) on YAG. Afterwards this
sample is annealed for one hour in the PLD chamber at 450 °C in an argon atmosphere. The next section describes the structural properties of the annealed sample.

4.5.1 X-ray diffractometry

2θ – ω scan and rocking curve

The measured 2θ – ω scan is shown in Fig. 4.13(a). The Pt (111) reflection is caused by scattering on growing crystallites along the (111) direction as described in Sec. 3.3.1 and is identical to the one for annealed Pt on YAG in Fig. 3.8(a). Compared to YIG thin films grown on YAG (cf. Fig. 4.2(a)) the YIG (444) reflex of the YAG/YIG/Pt bilayer has a lower intensity indicating a worse crystalline quality. Calculating the out-of-plane lattice constants for both reflections as described in Sec. 3.2.2 yields \( a_{\text{Pt}} = (0.3895 \pm 0.0070) \) nm and \( a_{\text{YIG}} = (1.2417 \pm 0.0233) \) nm. Both values match within their uncertainties to the literature ones [27, 28, 31].

The measured rocking curves in Fig. 4.13(b) reveal that Pt is still polycrystalline and show no low mosaic spread component as it is the case for annealed Pt thin films on YAG in Fig. 4.7(b). This could be caused by the larger lattice mismatch between YIG and Pt compared to YAG and Pt (see Sec. 2.2). The rocking curve belonging to the YIG (444) reflex only shows a very high mosaic spread of the corresponding lattice planes and no small mosaic spread component compared to YIG on YAG in Fig. 4.3(a). Furthermore, it seems that also YIG is polycrystalline with some larger (111)-orientated crystallites causing the small peak in the 2θ – ω scan.

Reflectometry and simulation

Before the annealing of the bilayer a reflectometry is measured. Then the sample is introduced again into the UHV cluster and annealed for one hour at 450 °C in the PLD chamber. Afterwards another reflectometry is measured. Both received reflectometries are shown in Fig. 4.14(a) revealing the influence of the reheating process on the YIG and Pt thin films.

The corresponding simulations in Fig. 4.14(b) are both able to fit the measured reflectometries using the ideal system YAG/YIG/Pt. The first surprising result is the thickness of Pt before the annealing process ((6.3 ± 0.1) nm) since a value comparable to (6.9 ± 0.1) nm is expected (cf. Fig. 3.9(b)). Both values do not match within the given uncertainty. This difference can either
be caused by additional measurement inaccuracies in the EVAP, which have not been taken into account in the given errors, or a reduced Pt growth rate on YIG compared to GGG|Pt.

The main difference in both simulations is the increasing thickness of Pt and the decreasing one of YIG, which can not be explained within the given errors. Since no PtO has to be assumed to improve the simulation it could be that Pt diffuses into YIG. Because of the much higher density of Pt compared to YIG (see Sec. 3.4.1), Pt dominates in the intermixing region causing the measured thickness change in the reflectometry. However, further experiments are necessary to examine the interface between YIG and Pt in more detail. The next section discusses the surface and magnetic properties of the YAG|YIG|Pt bilayer.

4.5.2 Atomic force microscopy and SQUID magnetometry

Because the time outside of the UHV cluster between the deposition of Pt in the EVAP and the annealing process is tried to minimize, only a x-ray reflectometry is carried out and no surface topography using atomic force microscopy is recorded. However, an AFM picture is taken after the annealing process. This results in a surface roughness of $R_{RMS} = (0.3 \pm 0.1) \text{ nm}$ and $(0.4 \pm 0.1) \text{ nm}$ afterwards.

Using SQUID magnetometry for the measurement of the magnetic hysteresis of the annealed YAG|YIG|Pt bilayer gives a saturation magnetization $M_S = (101.4 \pm 12.8) \text{ kA m}^{-1}$ which is identical to the one for YIG thin films on YAG in Fig. 4.6 within the uncertainty. Also a very small coercive field $\mu_0 H_C = (0.8 \pm 0.1) \text{ mT}$ is measured. This shows that the additional deposition of Pt and the following annealing process have no significant influence on the ferrimagnetic ordering in the YIG thin film.

4.5.3 Conclusion

The investigation of the annealing process of the YAG|YIG|Pt bilayer reveals that there is a detectable difference compared to the annealing of Pt thin films on YAG at 450°C in Sec. 4.3.
As soon as a YIG layer is deposited between the substrate YAG and the Pt thin film the surface and probably interface quality as well as the thin film thicknesses change during the annealing process. This could be caused by the different lattice mismatches as mentioned in Sec. 4.5.1. However, the ferrimagnetic behavior of YIG is not influenced by the overlying Pt thin film. Since a substrate temperature has been found where both YIG layers show a ferrimagnetic ordering the fabrication of a trilayer on YAG is described in the next section.

### 4.6 Trilayer on $\text{Y}_3\text{Al}_5\text{O}_{12}$

Finally, a YIG|Pt|YIG trilayer is fabricated on the substrate YAG using a substrate temperature of 450 °C for the growth of both YIG layers in an argon atmosphere in the PLD chamber as shown in Fig. 3.1(a). For Pt once again a thickness of 10 nm is set in the EVAP. The next section discusses the structural properties of the fabricated trilayer using x-ray diffractometry.

#### 4.6.1 X-ray diffractometry

**2θ − ω scan and rocking curve**

The measured $2\theta$ − $\omega$ scan of the trilayer is shown Fig. 4.15(a). The behavior of the Pt (111) reflection is similar to the behavior of annealed Pt thin films as described in Sec. 3.3.1. The YIG (444) reflection is caused by the first YIG layer (YIG1) (cf. Fig. 3.1(a) as well as Fig. 4.2(a)) since the second YIG layer (YIG2) does not have a crystalline structure according to Fig. 4.10(a). Calculating the out-of-plane lattice constants as described in Sec. 3.2.2 yields $a_{\text{Pt}} = (0.3863 \pm 0.0146)$ nm and $a_{\text{YIG}} = (1.2432 \pm 0.0128)$ nm. Again both values are in agreement within their uncertainty to the corresponding literature values [27, 28, 31].

The rocking curve of the YIG (444) reflex in Fig. 4.15(b) only contains a high mosaic spread component and confirms the worse crystalline quality of the YIG1 layer grown on YAG at 450 °C. Compared to YIG thin films on YAG in Fig. 4.3(a) the low mosaic spread component is missing. The rocking curve of the Pt (111) reflex is similar to the one in Fig. 4.13(b) showing that Pt is still polycrystalline.
Reflectometry and simulation

The reflectometry and corresponding simulations of the YIG|Pt|YIG trilayer on YAG are shown in Fig. 4.16. Compared to Fig. 4.11(a) the Pt thickness fringes vanish very fast in Fig. 4.16(a), which could be caused by very rough interfaces between the different layers or a rough surface of the trilayer. For 2θ angles larger than 2.2° only YIG thickness fringes remain.

Trying to simulate the measured reflectometry as shown in Fig. 4.16(b) reveals that the ideal heterostructure YIG|Pt|YIG is not able do describe the Pt thickness fringes. A first improvement is, as described in Sec. 3.4.1, the assumption of PtO between the two YIG layers of the trilayer. But even this simulation is not able to correctly fit the second larger oscillation. Here the combination of Pt and PtO between both YIG layers improves the fit quality. The system YAG|YIG1|Pt|PtO|YIG2 is used because, as shown in Fig. 4.14(b), the influence of the reheating between YIG1 and Pt can still be fitted using the ideal simulation. However, non of the three simulations is able to fit the whole reflectometry as it is possible for the trilayer on GGG in Fig. 3.16(b). Therefore, additional experiments are necessary to get a better insight into the interfaces and, thus, to improve the simulations. The surface and magnetic properties of the trilayer are discussed in the next section.

4.6.2 Atomic force microscopy and SQUID magnetometry

The measured surface topography of the trilayer using atomic force microscopy is shown in Fig. 4.17(a). The determined surface roughness of 2.43 nm matches with the value for the YIG2 layer obtained by x-ray reflectometry in Fig. 4.16(b). Furthermore, no facets are visible. The roughness is greater than in Fig. 4.12(a) for the inverse bilayer indicating that heating the substrate YAG twice to a temperature of 450 °C influences the YIG and Pt thin film inter- and surfaces additionally.
4.6 Trilayer on $Y_3Al_5O_{12}$

Figure 4.17: (a) Surface topography of the trilayer on YAG. (b) Magnetic hysteresis measured at 300K. For the calculation of the magnetization $M$ according to Eq. (A.3) the total magnetic moment $m$ of the sample is divided by the added volume of both YIG layers. The volume for one YIG thin film is calculated with Eq. (A.4) using the determined thicknesses of Fig. 4.16(b). More details are discussed in the text.

The measured magnetic hysteresis of the trilayer on YAG using SQUID magnetometry is shown in Fig. 4.17(b), which is clearly different compared to Fig. 3.17(b) for the trilayer on GGG. The magnetization is calculated by using the total volume of both YIG layers. The measured hysteresis can be divided into two separate hysteresis curves. One with a very small coercive field of $(72.2 \pm 1.0)$ mT and another one with a surprisingly high coercive field of $(1.2 \pm 0.1)$ T. These extracted values are calculated for each hysteresis curve as described in Sec. 3.2.4. The same applies for the saturation magnetization $M_S$ of the complete hysteresis curve. The value of $M_S = (121.2 \pm 3.7)$ kA m$^{-1}$ is smaller than the literature value of bulk YIG [25]. Since this saturation magnetization only slightly differs to the one of YAG|YIG in Fig. 4.6 and YAG|Pt|YIG in Fig. 4.12(b) the used calculation method for the magnetization $M$ by adding the YIG volumes seems to be justified.

The resulting question is which hysteresis curve is caused by which YIG layer in the trilayer YAG|YIG1|Pt|YIG2. Since YIG grown on Pt already shows a larger coercive field (cf. Fig. 4.12(b)) the huge coercive field of 1.2 T could be caused by the second YIG layer (YIG2). Interestingly also the first YIG layer (YIG1) gets magnetic harder with a coercive field of 72.2 mT compared to Fig. 4.6. However, further experiments are necessary to definitely assign the measured hysteresis curve to the two YIG layers.

4.6.3 Conclusion

A trilayer on YAG with two different adjustable magnetizations and a still conducting Pt spacer according to Ref. [44] can be successfully fabricated using a growth temperature of 450 °C for both YIG layers. Surprisingly one YIG thin film has a huge coercive field of 1.2 T indicating a hard magnetic behavior.
Chapter 5

Summary and outlook

The spin magnetoresistance (SMR) is one of the most intriguing property in the field of spintronics. In this subject area the fabrication of trilayers consisting of Y$_3$Fe$_5$O$_{12}$ (YIG) and Pt is the next milestone enhancing the MR effect and introducing an additional degree of freedom.

In this thesis YIG|Pt|YIG trilayers were made on Gd$_3$Ga$_5$O$_{12}$ (GGG) and Y$_3$Al$_5$O$_{12}$ (YAG) substrates. To get a better insight of the physical properties of each component of these trilayers, single YIG and Pt thin films as well as Pt|YIG and YIG|Pt bilayers were fabricated separately. The investigations of the structural, surface and magnetic properties of these samples were carried out by x-ray diffractometry, atomic force microscopy and SQUID magnetometry.

Trying to find consistent growth parameters for the fabrication of YIG|Pt|YIG trilayers on the substrate GGG has shown no success, since the Pt thin films exhibit a very high surface roughness for annealing temperatures larger than 350 $^\circ$C. However, at a substrate temperature as low as 350 $^\circ$C the second YIG thin film grown on a polycrystalline Pt layer is not ferrimagnetic at room temperature. Furthermore, (110)-facets are visible in the surface topography for all thin films on GGG with an annealing or deposition temperature larger than 350 $^\circ$C.

Using YAG as the substrate material the temperature where facets starts to form is shifted to 450 $^\circ$C probably due to a different surface energy of YAG compared to GGG. The single YIG and Pt thin films as well as the Pt|YIG and YIG|Pt bilayers fabricated at this temperature in argon exhibit excellent structural and magnetic properties. Therefore, a YIG|Pt|YIG could be realized on a YAG substrate using 450$^\circ$C as the deposition temperature for both YIG layers. The magnetic hysteresis recorded at 300 K shows a separate switching of both YIG layers with a about 7 nm thick Pt layer. However, one YIG layer has an unexpected high coercive field of 1.2 T indicating a hard magnetic behavior probably due to a high density of defects. Notwithstanding, the fabricated trilayer demonstrates the possibility to use YIG|Pt|YIG trilayers as a spin valve structure as discussed in chapter 2.

In all simulations of the x-ray reflectometries of samples containing annealed Pt thin films, a layer with a reduced density compared to Pt has to be inserted in the assumed heterostructure. This layer could be caused by oxidation of Pt or intermixing of YIG and Pt during the PLD process. Therefore, a reduction of the fluency at the target during the growth of the second YIG layer, which decreases the energy of the particle in the plasma plume, might lead to a lower intermixing of YIG and Pt. This could also be achieved by varying the pressure of argon in
the PLD chamber. Therefore, the examination of YIG thin films deposited on polycrystalline Pt layers with different growth parameters is mandatory to realize a good interface quality in future trilayers.

For a trilayer on YAG a consistent substrate temperature of 450 °C for both YIG layers has been found. However, the fabricated trilayer surprisingly shows a magnetic hysteresis, which contains a component with a high coercive field. To investigate this magnetic behavior in more detail, further SQUID magnetometry measurements must be carried out as a function of temperature and orientation of the external magnetic field direction. Moreover, additional trilayers with different Pt thicknesses have to be fabricated to test whether this magnetic behavior only occurs for this individual sample.
Appendix A

Experimental methods

A.1 Sample fabrication

A.1.1 Ultra high vacuum cluster

All samples are fabricated in chambers which are part of an ultra high vacuum (UHV) cluster as shown in Fig. A.1 with a bare pressure of about $10^{-9}$ mbar.

![Ultra high vacuum (UHV) cluster](image)

Figure A.1: Ultra high vacuum (UHV) cluster used for the thin film fabrication. The samples are introduced into the UHV at the load-lock and can be transferred into the pulsed laser deposition (PLD) or electron beam evaporation (EVAP) chamber over a transfer arm. All chambers are separated over plate valves (yellow rectangles) from the transfer chamber. The additional sputter chamber is not used.

The substrate (GGG (111) or YAG (111)) is pre-sputtered [45] with a 180 nm thick Pt film on its backside to guarantee a better absorption of the infrared laser light in the pulsed laser deposition (PLD) chamber during the heating process (see next Sec. A.1.2). After cleaning the substrate with isopropanol under the microscope and purging the sample holder with aceton and isopropanol both are introduced into the UHV at the load-lock. Over a transfer arm the sample can be transferred into the PLD or electron beam evaporation (EVAP) chamber depending on the fabrication steps according to Fig. 3.1. The UHV cluster enables the possibility to produce all layers in situ (without breaking the vacuum) to avoid contamination of the surface leading to a good interface quality between different layers.
A.1.2 Pulsed laser deposition

The YIG thin films are grown epitaxially using pulsed laser deposition (PLD) in the PLD chamber of the UHV cluster in Fig. A.1. The basic components are schematically shown in Fig. A.2.

![Figure A.2: Functional principle of pulsed laser deposition. The pulsed KrF excimer laser is focused by a telescope onto a rotated target. The emerging plasma plume reaches a substrate that is heated by an infrared laser allowing the epitaxial and stoichiometric growth of the thin film. The pyrometer measures the actual temperature of the substrate and the shutter protects the substrate during the cleaning process of the target. For the growth of a thin film a gas atmosphere is needed in the chamber to thermalize the hot particles coming from the plasma plume. The in situ analysis method reflection high-energy electron diffraction (RHEED) is illustrated by the incident e-beam and the fluorescence screen visualizing the scattered electrons.](image)

After the sample has been transferred into the chamber an argon atmosphere of 25 µbar pressure is set up. Then the substrate is heated up to the desired temperature with an infrared heating laser [46]. The reached substrate temperature is measured from the back and the front with two pyrometers. For the growth of YIG thin films a pulsed ultraviolet KrF excimer laser [47] is focused by a telescope onto a rotated stoichiometric, polycrystalline YIG target. Before the growth starts the shutter is closed and the YIG target is cleaned with a series of 2000 pulses. A laser fluency of 2 J cm\(^{-2}\) is used. The gas atmosphere is needed to thermalize the hot particles coming from the emerging plasma plume from one laser pulse and to avoid resputtering of the sample. During the growth a series of \(40 \times 285 = 11400\) laser pulses is fired at the YIG target with a relaxation time of 25 s between of them to reach a certain thickness. The repetition rate of the laser pulses is set to 10 Hz. Furthermore, the plasma plume is supposed to contain stoichiometric YIG and, therefore, enabling the deposition of a high-crystalline YIG thin film on (111)-orientated substrates (GGG or YAG). After the growth of YIG the sample is cooled down and the PLD chamber is evacuated again.

More information about the non-equilibrium growth of YIG with PLD and the in situ analysis method reflection high-energy electron diffraction (RHEED) for controlling the layer growth as well as the different growth modes for epitaxial films can be found in Refs. [39, 41, 48–50].

A.1.3 Electron beam evaporation

Platinum (Pt) is deposited on the sample using electron beam evaporation (EVAP) at room temperature (300 K) as shown in Fig. A.3.

The Pt is placed in a high temperature resistant crucible and gets heated up by an electron beam. This beam origins from electrons emitted by a filament (current of 30 A), accelerated by an electrode (high voltage of 80 kV) and finally focused in a magnetic field \(B\) on the Pt placed in the crucible. The emission current of the beam is about 135 mA and leads to the evaporation of the Pt at a pressure of about \(5 \times 10^{-7}\) mbar in the EVAP chamber with a growth rate of about
A.2 Analysis methods

A.2.1 X-ray diffractometry

The 4-circle x-ray diffractometer D8 Discover from Bruker [51] is used for the analysis of the structural properties of the grown thin films. This technique measures the intensity of monochromatic x-rays which are diffracted by the sample.

The x-radiation is generated by a copper (Cu) x-ray tube with an applied voltage of 40 kV and a current of 40 mA. Then only the emitted Cu-K$_\alpha$ radiation is parallelized by a Göbel mirror, which suppresses the bremsstrahlung and the characteristic Cu-K$_\beta$ radiation and is reduced to the Cu-K$_\alpha_1$ line by a 4-fold Ge-monochromator with a wavelength of $\lambda = 0.154056$ nm as shown in Fig. A.4(a). Since additional aperture slots are inserted into the optical path, the beam has a line shaped profile in the x-direction. The intensity of the scattered x-rays is detected by a detector with an angle resolution of 0.0001°. More information about the named components can be found in the data sheets from Ref. [51].

The sample is hold by underpressure from the back onto a sample holder and can be turned around all three spatial directions ($\omega$, $\chi$ and $\varphi$) with an Eulerian cradle. Thereby $\omega$ is the angle

1.1 Å s$^{-1}$. The crucible has to be grounded to avoid a negative charging of the system which would influence the electron beam.

The set thickness of 10 nm for Pt is measured by an oscillating crystal through the resonance change caused by the increasing mass through the vaporized material. An electronic system controls the shutter. When the signal is received to start the deposition of Pt the shutter is opened and automatically closed when the final thickness is reached. The substrate is placed in a distance of about 40 cm above the crucible. Compared to the PLD the EVAP uses thermal evaporation of a metal (here Pt), which has the advantage that the atoms have much less energy compared to the one in the plasma plume and, thus, avoid boundary surface reactions with the substrate. Furthermore, no gas atmosphere is needed to thermalize them.

Due to losses caused by measuring uncertainties such as a different position of the oscillating crystal compared to the substrate, the actual deposited thickness of Pt is determined using x-ray diffractometry which is described in the next section. This technique also reveals that Pt growth polycrystalline in the EVAP [10].
Appendix A  Experimental methods

Figure A.4: (a) Monochromatic x-rays from the Cu-Kα line irradiate with an angle of ω relative to the x-y plane, are diffracted by the sample and then measured by a detector at an angle of 2θ relative to the incident x-ray beam. The sample can be turned around all three spatial directions with the Euler angles ω, χ and φ. (b) Ideal crystal in the upper part where the lattice plane normal is parallel to the surface normal. In real crystal exits a small tilt of the lattice planes (e.g. from polishing) leading to the angle α between both normals.

between the sample surface and the incoming x-ray beam. φ denotes the in-plane and χ the out-of-plane orientation of the sample. 2θ is the angle between the incoming and scattered direction of the x-rays reaching the detector. This angle can be calculated to $2\theta = 2 \cdot \omega + \alpha$ with α taking into account the in-plane tilted lattice planes as shown in Fig. A.4(b). This tilt has to be considered during the alignment as well as the out-of-plane tilt over the angle χ to maximize the intensity at the detector. The x-ray beam is centered in x- and y-direction onto the middle of the sample. In z-direction half the intensity passes the sample while the other half penetrates into it for an angle position of $\omega = 0^\circ$ and $2\theta = 0^\circ$.

Figure A.5: (a) Bragg equation in the real space. The incident x-ray beam $k_i$ with an incident angle $\omega$ is scattered at two different lattice planes with distance $d_{hkl}$. The detector is placed at an angle of $2\theta$ and measures constructive or destructive interference depending on the path difference $\Delta s$ between $k_i$ and the leaving x-ray beam $k_f$. The same situation is shown in (b) for the reciprocal space in the (111) direction since all used substrates are orientated along that direction. When the scattering vector $q$ is equal with a reciprocal lattice vector (represented by the (444) lattice point) constructive interference occurs. The green lines indicate the changes for $q$ with the different scan types.

The fundamental equation for x-ray diffractometry is the Bragg equation [52]

$$2d_{hkl} \sin \omega = \lambda \quad \text{(A.1)}$$

with the wavelength $\lambda$ of the x-ray beam, the incident angle $\omega$ (Bragg angle) and the lattice plane distance $d_{hkl}$ with the Miller indexes $h$, $k$ and $l$ as shown in Fig. A.5(a). Equation (A.1)
describes the constructive interference of the incident beam $k_i$ with the scattered beam $k_f$ when the path difference $\Delta s$ is a multiple of the wavelength $\lambda$. This condition is similar to the vectorial Laue equation $G_{hkl} = q$ when the scattering vector $q = k_f - k_i$ is identical with a reciprocal lattice vector $G_{hkl}$ [52].

For a cubic lattice structure the lattice plane distance $d_{hkl}$ can be calculated over the lattice constant $a$ with the Miller indexes $h$, $k$ and $l$ [52]:

$$d_{hkl} = \frac{a}{\sqrt{h^2 + k^2 + l^2}} \quad \text{(A.2)}$$

In the next paragraphs the used measurement methods are described, which yield intensity (from the detector in counts per second (cps)) versus the corresponding angle plots.

### Rocking curve ($\omega$ scan)

This scan type is sketched in the reciprocal space for the (111) direction in Fig. A.5(b) and serves for the detection of the lattice plane tilt angle $\alpha$ from Fig. A.4(b). Therefore, the detector is fixed at a $2\theta$ value, where a high intensity reflex is expected (e.g. the allowed (444) peak of the substrate GGG or YAG). Then $\omega$ is typically varied in the range of $[2\theta/2 - 1^\circ; 2\theta/2 + 1^\circ]$ to find the position with a maximal detectable intensity. In the reciprocal space the rocking curve is represented by a fixed scattering vector $q$ rotated on a circuit around the origin.

The rocking curve can also be used to detect the mosaic spread of a crystal due to tilted lattice planes and, therefore, judge the crystalline quality. The full width at half maximum (FWHM) of a peak measured with a $\omega$ scan is a quantity for the mosaic spread (with a low FWHM indicating a high crystalline quality) while the intensity of the corresponding component is proportional to the amount of contributing lattice planes. A detailed explanation of the mosaic spread is given in Ref. [53].

### $2\theta$ – $\omega$ scan

After the correct alignment and determination of the tilt angle $\alpha$ as well as the out-of-plane tilt $\chi$ the $2\theta$ – $\omega$ scan can be measured according to $2\theta = 2 \cdot \omega + \alpha$ with $2\theta \in [10^\circ; 130^\circ]$. In the reciprocal space the scattering vector $q$ only changes its absolute value in the (111) direction as shown in Fig. A.5(b) detecting all allowed peaks of the garnet structure, which can be calculated over the structure factor [52].

The $2\theta$ – $\omega$ scan serves to detect in general the crystalline growth of the thin films, impurity phases, Laue oscillations and the determination of the lattice plane distance in the (111) direction allowing the calculation of a lattice constant in the (111) direction (out-of-plane). Since for the garnet structure only the (444) and (888) reflex occur in the examined $2\theta$ angle area an undisturbed cubic lattice symmetry is supposed using Eq. (A.2). Otherwise, if more reflexes are available, the Nelson-Riley method [54] can be used to calculate the out-of-plane lattice constant with a higher precision.

For the detection of the in-plane lattice constant and determination of the crystal symmetry asymmetric reflexes, which are not lying in the (111) direction, have to be examined by reciprocal space maps (RSM) [55]. This method also allows the determination of strained or relaxed growth of the thin films. In the first case the film adapts the in-plane lattice constant of the substrate while in the second case the thin film reaches its own bulk lattice constant [56].

Because epitaxially grown thin films have a finite number of lattice planes minor maximums appear similar to the diffraction situation at a slot. These minor maximums are called Laue oscillations and can be detected in $2\theta$ – $\omega$ scans. The condition for the occurrence of these oscillations is the coherent growth of the lattice planes on the substrate, which is indicating a
Appendix A Experimental methods

high crystalline quality of the thin film. On the other hand, relaxed film layers or a high surface roughness prevent Laue oscillations [37].

Reflectometry

In this scan type the small angle x-ray scattering (2θ − ω scan with 2θ ∈ [0.5°; 5°]) is used to determine the thin film thickness and roughness. The x-rays are reflected at all occurring interfaces, at the air - sample surface or at the film - substrate surface and interfere with each other. The measured intensity profile contains maximums and minimums, so called Kiessig fringes [36] depending on the path difference of the interfering x-rays. This curve is fitted with a simulation of the corresponding layer system with the software LEPTOS provided from Bruker [57]. The first minimum in the reflectometry occurs at a critical angle 2θ_{critical} when there is no more total reflexion of the x-radiation on the topmost layer (all mediums are optical thinner for x-rays compared to air [36]).

The period of the Kiessig oscillations gives the layer thickness with an inverse proportionality to the sine of the period [36], analogous to the diffraction by a double slit where the slit width and gap can be calculated from the distance of the intensity maximums. The exponential drop of the reflectometry is a quantity for the surface roughness. Since the used x-radiation has a line shaped profile the roughness information is also limited into one dimension. Here the atomic force microscope provides much better information as described in the next section.

A.2.2 Atomic force microscopy

The probe-scanning (fixed sample) MFP-3D atomic force microscope (AFM) from Asylum research is used to analyze the surface quality of the thin films with a height resolution of 0.06 nm [58]. The basic principle of an AFM is sketched in Fig. A.6.

Figure A.6: Basic schematic of an AFM. The cantilever with a sharp ending tip oscillates near the sample surface in tapping mode and is driven by the z piezo which also controls the height. The x and y piezo move the probe around the sample surface. The bending of the cantilever is measured by the photodetector over the deflection of the laser beam. The controller calculates the feedback signal for the z piezo to keep the amplitude of the cantilever oscillation upright. All signals are evaluated with the PC.

A cantilever with a very sharp ending tip (also called probe) from Olympus [59] is driven near its resonance frequency. This operation mode is one of the oscillating modes of the AFM.
All available topographic modes are shown in Fig. A.7(a) with their corresponding operating regime in the force-distance curve. This curve can be calculated by monitoring the deflection of the cantilever when the z piezo is used to move the tip towards the sample surface. Here the intermittent contact mode (also called tapping mode) is used where the cantilever oscillates in the regime depicted by the gray area (repulsive and attractive forces). The advantage of this mode is the possibility of the tip to pass through the contamination layer due to the measurement in air as shown in Fig. A.7(b) and, therefore, can touch the actual sample surface.

Figure A.7: (a) Three different topographic modes of an AFM and their corresponding regime on the force-distance curve. The intermittent contact (tapping mode) and non-contact mode are the two oscillating modes. In the latter attractive and repulsive forces are acting on the cantilever during its oscillation. (b) Problem of a contamination layer for a measurement in air. In tapping mode the tip passes in and out of this layer (indicated by the two arrows) to touch the actual sample surface. (c) Change in the oscillation amplitude of the cantilever due to a step in the surface. The z piezo tries to keep it constant by adjusting the height of the cantilever above the sample.

The horizontal and vertical bending of the cantilever is measured over the deflection of the laser beam by the photodetector in Fig. A.6. In tapping mode the amplitude of the cantilever is used as the feedback signal and tried to be kept constant. Figure A.7(c) shows the effect of a step in the sample surface. The amplitude gets smaller and phase shifts occur relative to the driving frequency. These changes are readjusted by the z piezo to keep the set value for the amplitude upright (so called set point). During the measurement the amplitude, phase shift and z piezo signal are monitored. The additional height plot is calculated from the z piezo signal and is used in this work for further analysis.

To move the probe over the sample surface additional x and y piezos are used. All described components are evaluated on the PC with the control software IGOR Pro [60] resulting in two-dimensional image data. The analysis of these images (height plots) is done with the software WSXM [61] to extract the surface roughness over the root mean square value of the average surface height.

The scan parameters are an imaging area of $5 \mu m \times 5 \mu m$ with $512 \times 512$ scanlines and a scanspeed of about $20 \mu m s^{-1}$. The setpoint for the amplitude is about $0.6 V$. For better imaging results the AFM is protected by a vibration isolation (consisting of a platform, table and a surrounding cabinet). Further details about the AFM measurements and the other topographic modes as well as occurring difficulties during a scan (as drifts, a sticking tip or overmodulation signals) are described in [62]. There are also specified all necessary alignments before the start of a scan, several more scan parameters (e.g. the integral gain, drive amplitude or proportional gain) and details about the electronic feedback system.
A.2.3 SQUID magnetometry

The superconducting quantum interference device (SQUID) magnetometer MPMS from Quantum Design [63] is used to measure the magnetic moment $m$ of a sample to receive its magnetization $M$. Therefore, the magnetic background of the substrate (GGG or YAG) has to be subtracted and then the remaining magnetic moment is divided by the volume $V$ of the thin film to calculate the magnetization

$$M = \frac{m}{V}. \quad (A.3)$$

The sample is glued with a diamagnetic glue [64] in a straw as shown in Fig. A.8(a) and is moved up and down in the superconducting loop geometry (second-order gradiometer [65]). The sinusoidal movement of the specimen is called a reciprocating sample option (RSO) measurement. This special adjustment of the conducting loops minimizes the effect of noise through interfering fields or fluctuations. Furthermore, the straw is always moving within all four loops and, thus, creates no signal.

The whole scene shown in Fig. A.8(a) is surrounded by a helium cooled superconducting coil producing magnetic fields $\mu_0 H$ between $-7$ T and $7$ T. In addition, the sample space can be cooled within a temperature range of $1.8$ K to $400$ K. Here all measurements are done at $300$ K to record the magnetic moment $m$ versus the external magnetic field $\mu_0 H$ curves.

Through the magnetic field of the superconducting coil and the up-down movement of the sample a current is induced in the superconducting loops, which allows the measurement of the component of the magnetization into the direction of the external field. Therefore, the induced current is transformed by a radio frequency (RF) SQUID [66, 67] into a voltage as shown in Fig. A.8(b). This voltage curve is then fitted by a software from Quantum Design allowing the calculation of the magnetic moment $m$.

As mentioned above the thin film volume $V$ is needed to finally calculate the magnetization $M$. Therefore, the thin film surface area $A$ is determined using a microscope (5x objective) [68]. All the places are photographed where no thin film has grown on the substrate and their area is

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**Figure A.8:** (a) Second-order gradiometer that is formed out of a superconducting wire. The upper and bottom coils are single turn wound clockwise (-1) while the center coil comprises two turns wound counter-clockwise (+1). The sample is glued in the straw that moves up and down and induces a current in the gradiometer due to the external magnetic field $H$, which is transformed into a voltage by the SQUID. (b) Resulting voltage curve depending on the sample position.
calculated with the software ImageJ [69] (using the known conversion factor between pixels and millimeters for the 5x objective: 86 pixel $\approx 0.1$ mm) which is then subtracted from the substrate area of 25 mm$^2$. The thickness $d$ of the thin film can be extracted from a x-ray reflectometry measurements as described in Sec. A.2.1 leading to

$$ V = A \cdot d. \tag{A.4} $$

According to Quantum Design [63] the minimal measurable magnetic moment $m$ is about $10^{-8}$ emu. For the following calculation a minimal value of $m \approx 10^{-7}$ emu is supposed. Taking the magnetization of YIG of about $M \approx 150$ kA m$^{-1}$ and a thin film surface area of ca. $A \approx 23$ mm$^2$ the minimal thickness of the detectable magnetic layer is

$$ d = \frac{m}{A \cdot M} \approx 0.03 \text{ nm} \tag{A.5} $$

according to Eq. (A.3) and Eq. (A.4). This result shows the sensitivity of a SQUID magnetometer and its applicability in the detection of magnetic moments in thin film technology.
Appendix B

Sample view

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<tr>
<th>sample</th>
<th>layers</th>
<th>atmosphere</th>
<th>substrate temperature $T_s$</th>
<th>$d$ (nm)</th>
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<td>YIG</td>
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<td>Ar</td>
<td>550 °C annealing (1 h)</td>
<td>–</td>
</tr>
<tr>
<td>YY#44</td>
<td>YAG</td>
<td>YIG</td>
<td>Ar</td>
<td>350 °C</td>
</tr>
<tr>
<td>YY#46</td>
<td>YAG</td>
<td>YIG</td>
<td>O$_2$</td>
<td>500 °C</td>
</tr>
<tr>
<td>YY#49</td>
<td>YAG</td>
<td>YIG</td>
<td>Ar</td>
<td>450 °C</td>
</tr>
<tr>
<td>YY#52</td>
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<td>YIG</td>
<td>Ar</td>
<td>550 °C</td>
</tr>
<tr>
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<td>Pt</td>
<td>Ar</td>
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<tr>
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<td>Pt</td>
<td>Ar</td>
<td>550 °C annealing (1 h)</td>
</tr>
<tr>
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<td>Pt</td>
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<td>Ar</td>
</tr>
<tr>
<td>YY#51</td>
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<td>Pt</td>
<td>YIG</td>
</tr>
</tbody>
</table>

Table B.1: Sample view. Explanation is given in the text.

Table B.1 lists all fabricated samples. The used substrates GGG and YAG are orientated into
the (111) direction. The YIG growth parameters are a corresponding substrate temperature $T_S$ and gas atmosphere which always has a pressure of 25 µbar in the PLD chamber. The excimer laser fires $40 \times 285 = 11400$ pulses on the YIG target with a relaxation time of $t_r = 25\, s$ between two packages. One pulse series has a repetition rate of $f = 10\, Hz$ and the energy density at the YIG target is $\rho_L = 2\, J\, cm^{-2}$. Annealing means that the listed gas atmosphere is adjusted in the PLD chamber and the substrate is heated for the given time at the specific temperature. The listed thicknesses $d$ (in nanometers for the final sample) in one table line have the same order as the corresponding layer structure excluding the substrate. The dash $-$ indicates thicknesses that can not be extracted of the reflectometry simulations. The samples marked with a star $\star$ are not used for the data evaluation. Furthermore, the samples marked with a diamond $\blacklozenge$ are used by D. Irber for the measurement of the SMR effect [44].
References


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