Unconventional Hall Effect in Ultra-thin SrRuO$_3$ Films

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Unconventional Hall Effect in Ultra-thin SrRuO₃ Films

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Abstract

Ultra-thin films with different thicknesses of SrRuO₃ were grown epitaxially on SrTiO₃. The anomalous Hall effect (AHE) with additional peaks was observed in Hall resistivity measurements as a function of field and temperature in a 5 unit cell SrRuO₃ film without capping layer. The additional peak phase matches literature, whereas the actual resistivity size is found to be lower. The peaks could be explained by either a topological Hall effect (THE) caused by the presence of a skyrmion lattice or two phases of the AHE corresponding to different interfaces. If the additional effect is a THE, this study confirms the presence of a robust skyrmion phase in ultra-thin SrRuO₃ on SrTiO₃ without capping layer, while gating experiments indicate that the skyrmion size could be tuned by an electric field.
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Chapter 1

Introduction

Research in the field of spintronics has shown many advantages over conventional semiconductor data storage and processing. One of the main problems with the ever increasing amount of data is energy loss through dissipation in the form of heat. In spintronics the spin of electrons is used for data storage and processing instead of the electron’s charge. Due to their spin properties, ferromagnets can for example be used to create racetrack memories for fast data processing, in which domain walls function as databits [1]. Furthermore, superconductors were added to the field of spintronics to investigate opportunities for reducing the problem of energy dissipation and to utilize emergent phenomena at the interface of ferromagnets and superconductors in devices.

Over the last years a lot of interest has been shown in skyrmions after first observing them in non-centrosymmetric magnetic compounds [2, 3]. These vortex-like spin structures in ferromagnetic materials could bring many potential advantages to the field of spintronics due to their interesting topological properties [4, 5]. It was shown that skyrmions could be moved with a process similar to current-induced domain wall motion. As skyrmions require lower current densities than regular domain walls, are defect tolerant, and seem stable over a broad range of magnetic fields and temperatures, the way has been opened for ‘skyrmionics’ as an even more effective means of data storage and processing [6].

Recently, studies reveal indications of skyrmions in ultra-thin layers of the ferromagnetic oxide SrRuO$_3$ by means of transport measurements [7, 8], while these indications are simultaneously questioned by other models [9]. This work is a contribution to this discussion by showing important inconsistencies the previous literature and comparing it with our findings, questioning the assumption of the presence of skyrmions in SrRuO$_3$. 
Chapter 2

Theoretical Background

The essential background theory will be discussed as follows. First of all we will define the nature of skyrmions and explain their origin. Secondly, we will give a recap of the ordinary and anomalous Hall effect and explain the origin of the topological Hall effect.

2.1 Skyrmions

2.1.1 Definition

Skyrmions are topologically protected vortex-like spin-structures in a ferromagnet. They can be referred to as quasi-particles due to their stability [10]. Skyrmions are topologically protected meaning that they are stable due to their topology, i.e. their shape. To make this property more intuitive, we will start with a discussion of what skyrmions look like and what the differences are with regular magnetic vortices.

In a ferromagnet, the orientation of the spins of the electrons can be such as to construct different kinds of structures. The variety in structures is often due to a balancing of the ferromagnetic exchange interaction that tries to aligns neighbouring spins, and other interactions in the lattice such as anisotropy or local deformations. Examples of these structures are domain walls - the interfaces between two domains with different magnetisation directions - but also magnetic vortices that show spin orientations curling around a vortex core. Skyrmions are a specific kind of these magnetic vortices and in order to explain the difference, let us first turn to a one dimensional example as shown in Fig. 2.1. In Fig. 2.1(a) a 1d string of magnetic spins is shown, all oriented in the same direction thus forming one
magnetic domain. When we look at Fig. 2.1(b) we see three magnetic domains, separated by two domain walls in which the orientation is flipped 180°. Looking closer at these domain walls, the important point is that the spins in the second domain wall rotate in the opposite direction with respect to the situation in the first domain wall. However, in Fig. 2.1(c) in both domain walls the spins are rotated in the same direction. Now we can define the whirling number, also known as the skyrmion number \( S \), which is an index for the total rotation of the spin [11]. In the situation in Fig. 2.1(b), the whirling number \( S = 0 \), as a spin moving along the line undergoing two rotations in opposite directions add up to zero. Whereas, if the rotation in the second domain wall is in the same direction, we end up with a whirling number of \( S = 1 \), as shown in Fig. 2.1(c).

**Figure 2.1:** Different examples of the whirling number, depending on the orientation of the spin rotation in domain walls. Image taken from [11].

Now let us consider the two-dimensional case. Two 2D magnetic vortices are shown in Fig. 2.2. The vortex type shown in Fig. 2.2(a) is a so called double vortex. The type of vortex in Fig. 2.2(b) is a skyrmion. By applying the same logic as in the 1D case, let us draw an imaginary straight line from the vortex perimeter to the vortex core of the double vortex as indicated in Fig. 2.2(a). Along the line there are again two domain walls of 180°. As we move along the line towards the centre, we find that the rotations in the domain walls are opposite to each other in the same way as the situation in Fig. 2.1(b), showing that this vortex has whirling number \( S = 0 \) (extending the line to the vortex perimeter doesn’t change this number). Now compare this to a diameter line shown in Fig. 2.2(b). The spins in the two domain walls along this lines rotate in the
same direction, resulting in a whirling number of $S = 1$. Therefore, this structure can be referred to as a skyrmion. The property described here is what fundamentally defines skyrmions as a separate class of magnetic vortices.

![Image of Magnetic Vortices](image)

**Figure 2.2:** Magnetic vortices. Domain walls between the blue and red domains are illustrated with a white color. (a) Double vortex. (b) Skyrmionic vortex. Images taken from [12].

The rotation of the spins mentioned above, can appear in two directions. This gives rise to two different kinds of skyrmions, shown in Fig. 2.3. They are named Néel-type skyrmions for the skyrmion in Fig. 2.3(a) and Bloch-type skyrmions for the skyrmion in Fig. 2.3(b), analogous to the names for the different types of domain walls [13]. The two types of skyrmions share the same fundamental properties, and the interaction giving rise to skyrmions generally determines the type present [14].

![Image of Two Types of Skyrmions](image)

**Figure 2.3:** Two types of skyrmions. (a) Néel-type skyrmion. (b) Bloch-type skyrmion. Image taken from [4].
Now we understand the fundamental topology of a skyrmion, one might wonder how a skyrmion’s shape gives rise to the property of being topologically protected. We can intuitively understand this by imagining a magnetic field that is applied in the out of plane in the direction, which is the direction in which the outermost spins point in all examples. In the 1D example, the helix with $S = 0$ will slowly untie itself with increasing magnetic field in the upward direction, whereas the helix with $S = 1$ will knot itself even more. Similarly applying a magnetic field in the z-direction Fig. 2.2(a) will continuously untie the vortex, whereas applying a field in the negative z direction in Fig. 2.2(a) will create a two dimensional knot that can only be untied discontinuously by applying a very high magnetic field or going to higher temperatures. This is why skyrmions are per definition robust entities. As their geometrical structure determines this robustness, skyrmions are called topologically protected [15].

Finally, it has to be mentioned that the three dimensional shape of a skyrmion extends in a cylinder shape in the z-direction, as shown in Fig. 2.4.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{skyrmion_lattice.png}
\caption{A visualisation of a 3D skyrmion lattice. Skyrmions are fundamentally two-dimensional entities, but can be enlarged in the out of plane direction forming a cylindrical shape. Image taken from [8].}
\end{figure}
2.1 Skyrmions

2.1.2 Origin

In this section we consider a ferromagnetic material in the vicinity of its interface. The origin of skyrmions in such a system lies in the Dzyaloshinskii-Moriya interaction (DMI) that competes locally with the ferromagnetic exchange interaction. When inversion symmetry is broken at the film interface, two neighbouring magnetic spins will couple with a third atom which has large spin-orbit coupling (SOC) [4]. This gives rise to a DMI vector \( \mathbf{D}_{12} \), resulting from the DMI between two spins \( S_1 \) and \( S_2 \) defined by the DM-Hamiltonian,

\[
H_{DM} = -\mathbf{D}_{12} \cdot (S_1 \times S_2).
\] (2.1)

The DMI rotates the first spin around the DMI vector with respect to the second spin. This interaction is visualised in Fig. 2.5. The direction of the DMI vector determines which of the different types of skyrmions described in Section 2.1.1 is formed. It has been shown that in ultra-thin films Néel-type skyrmions are formed [4, 7]. Inversion symmetry breaking can also be an intrinsic property of the lattice of a material, causing a DMI of which the details are not discussed here as we are mainly interested in inversion symmetry breaking at interfaces of films.

The other key ingredient for the existence of skyrmions is the ferromagnetic exchange interaction. As the energy is minimized between the exchange interaction that tries to align neighbouring spins and the DMI vector with a contribution perpendicular to the plane in which the spins are aligned, the favourable energy state is a state where the magnetic spins form skyrmions, called a skyrmion lattice. The size of skyrmions in this lattice is determined by the ratio between the DMI and exchange interactions. A higher ratio rotates the spins faster, resulting in a smaller skyrmion sizes.

![Visualisation of the origin of the DMI](image)

**Figure 2.5:** Visualisation of the origin of the DMI. Two neighbouring spins couple to a third atom with large spin-orbit coupling. Image taken from [4].
2.2 Hall effects

An indication for the presence of skyrmions in thin films is the topological Hall effect (THE). Given that this effect is both mixed with the ordinary Hall effect (OHE) and the anomalous Hall effect (AHE), we will first turn to a recap of these effects before introducing the THE.

2.2.1 Ordinary Hall effect

When a current $I$ is applied through a wire of thickness $t$, width $w$, and length $L$, a magnetic field $B$ applied perpendicular to the wire results in a Lorentz force on the electrons in the wire. This pushes the electrons to one side of the wire, giving rise to an electric field $\xi$, that balances the Lorentz force. The size of the Hall voltage $V_H$ that can be measured depends on the applied field, the wire thickness, the applied current, but also on the charge carrier properties of the material. To describe these material dependent properties, a Hall coefficient is defined,

$$R_H = \frac{\xi}{J B} \quad (2.2)$$

where $J$ is the applied current density. A schematic drawing of a typical Hall effect measurement setup is shown in Fig. 2.6.

![Figure 2.6: A Hall effect measurement setup.](image)

2.2.2 Anomalous Hall effect

For ferromagnets as well as for paramagnets in a magnetic field there is an extra contribution to the Hall effect that is directly dependent on the magnetisation of the material [16]. This effect is called the anomalous Hall effect (AHE), also referred to as the extraordinary Hall effect. The origin of this strongly temperature dependent effect is found in a number of mechanisms related to spin-orbit coupling. Two of these mechanisms are intrinsic effects due to scattering of charge carriers and depend on the spin,
and are called skew-scattering and side-jump. The other contribution to the AHE is due to intrinsic effects related to a Berry phase in momentum space due to the curvature of occupied nontrivial band structures at the Fermi level [9, 16, 17]. Furthermore, the AHE is linearly dependent on the magnetisation $M$, so follows a similar hysteresis loop in $\rho(H)$ as $M(H)$.

### 2.2.3 Topological Hall effect

The topological Hall effect (THE) is the direct contribution due to a spatially varying magnetisation which subtends a solid angle. Skyrmions have this intrinsically varying magnetisation as a result of their shape. When applying a current through a wire in which a skyrmion lattice exists, the spins of the electrons of the current will interact with the skyrmion via the spin-torque interaction. This moves the skyrmions in the direction of the current in a process similar to current induced domain wall motion [4]. This process is shown in Fig. 2.7. Due to the magnetic structure of the skyrmion, an emergent magnetic field arises which induces a Lorentz force on the passing current, deflecting the current to one side of the wire [18]. This effect is the THE, because due to the topology of the skyrmion lattice, an extra contribution to the Hall effect is introduced [18]. Additionally, as a result of the so called topological Magnus force due to the topological charge, or whirling number as discussed in Section 2.1.1, the skyrmion gains a transverse velocity component in the opposite direction to the Lorentz force which is acting on the current. This is called the skyrmion Hall effect [19] and is also illustrated in Fig. 2.7.

![Figure 2.7: An illustration of the topological and skyrmion Hall effect. Image taken from [18].](image-url)
Literature Review

Recent developments in literature are discussed considering the possible origin of skyrmions in SrRuO$_3$. Additionally, literature views on the growth of SrRuO$_3$ are reviewed and eventually, recent measurements of the topological Hall effect in SrRuO$_3$ are discussed.

3.1 Skyrmions in SrRuO$_3$

Recently it was shown that the perovskite oxide SrRuO$_3$ is a material that shows indications for hosting skyrmions [7, 8]. SrRuO$_3$ is an oxide in the class of the ruthenates that shows many interesting properties, such as itinerant ferromagnetism and unusual transport behaviour [20]. But the most important properties for our purposes are that SrRuO$_3$ is both a ferromagnet and exhibits strong intrinsic spin-orbit coupling, providing the basic ingredients for skyrmion formation [4]. As described in Section 2.1.2 the strength of the DMI has to be sufficient to compete with the ferromagnetic exchange interaction to create skyrmions. To be able to make the contribution of the DMI compete with the exchange energy, it is shown that ultra-thin magnetic films are needed to increase the contribution of inversion symmetry breaking at the interface [4, 7]. In the case of skyrmions in SrRuO$_3$, the thickness of the films for which indications of skyrmions were observed are mainly 4 and 5 unit cells (u.c.) of SrRuO$_3$, corresponding to 1.6 - 2.0 nm [7, 8]. The estimated size of skyrmions in these types of lattices is about 10 nm [7]. Initially, it was thought the intrinsic spin-orbit coupling of SrRuO$_3$ was too small to be able to give rise to the DMI necessary for creating skyrmion structures. When the first indications for a skyrmion phase were found in SrRuO$_3$, a capping layer of SrIrO$_3$ of a few
unit cells was provided in order to enhance the spin-orbit coupling [7]. However, it has since been shown that the same phase can be observed in ultra-thin SrRuO$_3$ without this capping layer. In this last study, the phase in the $HT$-plane was found to be larger than in other materials [3, 21]. The phase diagram is shown in Fig. 3.1, where the phase can clearly be seen up to 80 K and within a magnetic field range of about 1 T.

![Phase diagram of 5 u.c. of SrRuO$_3$. A ferromagnetic (FM), skyrmion (Sk) and spin-polarised phase (SP) are shown. The color map represents Hall resistivity values. Image taken from [8].](image)

**Figure 3.1:** The phase diagram of 5 u.c. of SrRuO$_3$. A ferromagnetic (FM), skyrmion (Sk) and spin-polarised phase (SP) are shown. The color map represents Hall resistivity values. Image taken from [8].

A common method for finding indications of the presence of skyrmions is by measuring the topological Hall effect (THE) by means of transport measurements [7, 17, 22, 23]. Another method is Lorentz transmission electron microscopy (LTEM) which gives spatial resolution and a more direct probe [21, 24, 25]. In this work we will focus on the THE.
3.2 Growth of SrRuO$_3$

A commonly used substrate for the growth of epitaxial thin films of SrRuO$_3$ is single crystal SrTiO$_3$ as its lattice parameters match closely as to ensure good quality growth. A lattice mismatch will generally increase the strain and thereby affect the properties of the material. A widely-used growth method is pulsed laser deposition (PLD), which will be described in Section 4.1.2. Furthermore, an important factor for successful growth of SrRuO$_3$ has been shown to be the single termination of the SrTiO$_3$ substrates [20]. This means the top layer of the SrTiO$_3$ has to consist of either TiO$_2$ or SrO, as the nucleation rate seems to vary on these terminations [20]. A common method for making SrTiO$_3$ single-terminated is the HF-treatment which provides very good quality single TiO$_2$ termination [20, 26, 27]. The surface of the TiO$_2$-terminated substrates is shown in Fig. 3.2(a). The substrate miscut is shown by the steps on the surface that are of unit cell height. In the growth process the step edges work as nucleation sites for a new layer to grow, enabling step flow growth [20]. For the best quality of growth literature recommends heating the substrate to 600°C - 700°C under an oxygen pressure of the order of 10 Pa during growth. To get rid of defects post-annealing in oxygen is recommended [20]. An example of what a nicely grown layer of SrRuO$_3$ looks like is shown in Fig. 3.2(b).

![Figure 3.2](image.jpg)

**Figure 3.2:** (a) Smooth surface of TiO$_2$-terminated SrTiO$_3$ substrate. (b) Example of smooth step-flow growth of 30 nm of SrRuO$_3$. (c) Trenched growth due to mixed termination of the substrate. Images taken from [20].

Furthermore, it was shown that due to the diffusion of SrO to the surface, SrTiO$_3$ can become mixed-terminated during annealing [20]. Due to the difference in interfacial energy between the two termination types, SrRuO$_3$ seems to grow faster on the SrO-terminated areas than on the TiO$_2$ areas [20]. Instead of a good quality film, this results in the growth of trenches, as shown in Fig. 3.2(c).
3.3 Hall measurements in SrRuO$_3$

As discussed in the previous chapter, when performing a Hall measurement on a skyrmion lattice, there will be three contributions, an ordinary, anomalous and topological contribution in the presence of skyrmions. This results in the total Hall effect [17],

$$\rho_{xy} = \rho_{xy}^O + \rho_{xy}^A + \rho_{xy}^T.$$  \hspace{1cm} (3.1)

Recently, Hall measurements have been performed in ultra-thin films of SrRuO$_3$ for thicknesses of 4, 5, 6 and 7 unit cells with a capping layer of SrIrO$_3$ to enhance SOC [7], as well as in just an ultra-thin film of SrRuO$_3$ for the same thicknesses [8]. The results from this last study is shown in Fig. 3.3.

**Figure 3.3:** Hall resistivity versus out-of-plane external field measured in ultra-thin films of SrRuO$_3$ for 4, 5, 6 and 7 u.c. thicknesses for different temperatures. The OHE term is substracted. The AHE can clearly be observed as hysteresis in all thicknesses. What is expected to be the THE is observed as an additional contribution near the coercive field for 4 and 5 u.c. thicknesses. Image taken from [8].

In these measurements, the field is swept from negative to positive and backwards, while measuring the transverse (Hall) resistance over a Hall bar. This data shown here is corrected for the linear OHE. The hysteresis behaviour is due to the AHE, which can clearly be seen in the graphs for
the 6 and 7 unit cell thicknesses. The extra peak present in the graphs for the 4 and 5 u.c. thicknesses was predicted to be due to the THE. In the domain of the applied field and temperature where this peak is observed, skyrmions are expected to be present. The phase diagram mentioned in Section 3.1 was derived from this data.

As both the AHE and the THE can give contributions to the Hall effect, they can be hard to separate from each other. The observation that AHE is both positive and negative, whereas the THE is only found to be positive was argued to be an indication that the origins of the two processes are truly separate [7, 8]. The position of the additional peaks always seem to be at the coercive field, which indicates that the THE could be enabled by the magnetisation reversal process [7].
3.4 Alternative explanation of the topological Hall effect

Very recently, another theory on explaining the effect priorly ascribed to the THE was proposed [9]. This view states that the observed peak of the THE is actually a result of a superposition of two contributions of the AHE from two separate spin-polarised conduction channels corresponding to the two different interfaces of the SrRuO$_3$. These interfaces deform the band structure of SrRuO$_3$ in different ways, giving rise to two different contributions of the AHE with different temperature and field dependences. This model was based on experimental results of symmetric heterostructures of SrTiO$_3$/SrRuO$_3$/SrTiO$_3$ and SrIrO$_3$/SrRuO$_3$/SrIrO$_3$ and asymmetric heterostructures of SrTiO$_3$/SrRuO$_3$/SrIrO$_3$. A collection of Hall measurements performed on these heterostructures are shown in Fig. 3.4.

![Figure 3.4](image)

Figure 3.4: Hall resistance of asymmetric SrTiO$_3$/SrRuO$_3$/SrIrO$_3$ and symmetric SrTiO$_3$/SrRuO$_3$/SrTiO$_3$ heterostructures. All SrRuO$_3$ layers have a thickness of 4 u.c. and SrIrO$_3$ layers have thicknesses of 2 u.c. Image taken from [9].

As shown in Fig. 3.4, the extra peak that is observed around the coercive field is only present in the asymmetric structure. It was argued that the peak arises only when there are two phases due to two different interfaces, while symmetric structures only give one phase as they only have one kind of interface [9]. To further demonstrate this, measurements have been compared with a model. This model is illustrated in Fig. 3.5, where
the two contributions to the AHE are shown on the right, adding up to the figure on the left, matching the experimental data.

Figure 3.5: Illustration of two anomalous Hall channels. On the left: the experimental data fitted with the model. On the right: the separate two contributions that make up the data on the left according to the model. Both as a function of temperature and magnetic field. Image taken from [9].

According to the beforementioned views on how the THE arises in SrRuO$_3$, a symmetric heterostructure with SrIrO$_3$/SrRuO$_3$/SrIrO$_3$ was expected to enhance the SOC of the SrRuO$_3$ even more, which should cause an even stronger DMI resulting in a larger THE [9]. However, in these heterostructures no peaks at the coercive field were observed, supporting the more recent model opposed to earlier views [7, 8]. Furthermore, it was argued that the fact that the peaks are specifically present during sign switching of the AHE, as can be seen in Fig. 3.4, supports this view. On the other hand, in the experiments without a capping layer, the peaks don’t seem to disappear at lower temperatures where there is no sign changing of the AHE [8]. Additionally, in the study with capping layer the data for 4 u.c. of SrRuO$_3$ show an increasing THE peak with decreasing temperature without any AHE sign change present [7]. Thus, the current debate on the origin of these effects indicate that the exact dynamics are not yet fully understood.
3.5 Electrical tuning of Hall effects

It was shown that both the peaks ascribed to the THE and the AHE could be tuned by applying an electric field in the vicinity of the film [28]. This was done by backgating the sample, using the SrTiO$_3$ substrate as a gate dielectric. An example of the change in AHE as a result of an applied electric field is shown in Fig. 3.6, where the difference of the size of the AHE can be clearly seen between an applied gate voltage of -180 V and 200 V. A similar relative change was seen in the THE [28]. The effects in tuning the THE and AHE were only found in samples where a layer of 2 u.c. SrIrO$_3$ was placed between the SrRuO$_3$ and SrTiO$_3$ substrate to enhance the SOC. In SrRuO$_3$ in these cases was always 5 u.c. thick.

![Figure 3.6: Hall resistivity data under gating dependent of the stacking order. These measurements were performed at 2 K. The OHE is substracted. Image taken from [28].](image)

The fact that no THE peaks are observed in the SrRuO$_3$ on SrTiO$_3$ without SrIrO$_3$ stack is in contradiction to the findings of [8], raising questions about the quality of the SrRuO$_3$ growth in the used sample, especially as no TiO$_2$ termination for the SrTiO$_3$ substrates was mentioned [7]. Moreover, it was reasoned that the modulations in the AHE and THE peaks was primarily the effect of probing the SOC, instead of primarily the charge carrier density. According to this view the AHE and THE are seperate effects, where the AHE is tuned as the SOC changes the bandstructure of SrRuO$_3$. The THE is affected as the SOC changes either the DMI (and thereby the skyrmion phase) in the skyrmion model, or curves momentum space resulting in a change in AHE in the alternative model.
Ultra-thin epitaxial SrRuO$_3$ was grown by pulsed laser deposition, and the structural and magnetic properties were characterised. Films were then patterned into Hall bars using photolithography in order to do Hall measurements.

4.1 Growth

4.1.1 Substrate preparation

The substrates used for the SrRuO$_3$ growth were Crystec TiO$_2$-terminated SrTiO$_3$ substrates. Initially, SrTiO$_3$ with mixed termination was used after which an annealing process was done to make the SrTiO$_3$ single-terminated. This annealing was done holding the substrate for 90 minutes at 950°C with a 10°C/min ramp rate and 60 sccm O$_2$ flow. Later, this step was skipped in favour commercial TiO$_2$-terminated SrTiO$_3$ substrates as this enables better growth quality.

4.1.2 Pulsed laser deposition

SrRuO$_3$ was deposited on the single-terminated SrTiO$_3$ substrates by pulsed laser deposition (PLD). The basic setup for PLD consists of a high power pulsed laser incident on a target of the material one wants to deposit. Upon hitting the target, the target material is ablated and a plume of plasma is formed with stoichiometry similar to the target [29]. This plume is incident perpendicular to the substrate surface. A schematic of the PLD setup is shown in Fig. 4.1. Furthermore, the sample is heated to allow for
allow for diffusion of atoms on the surface so they can rearrange into a stable crystal structure. Our substrates were heated to 600°C at a rate of 20°C/min. The whole process is performed in a vacuum chamber, while a oxygen pressure of between 11 and 13 Pa was used as a background gas to ensure optimal oxygen stoichiometry [29]. Before deposition, the vacuum chamber is kept at the deposition temperature to enable outgassing of contamination around the heater. The laser that was used is a KrF laser with an energy of 350-400 mJ and a pulse length of 25 ns. To ensure an uniform ablation of the target, a target rotator is used to spin the target during deposition. The target was pre-ablated for 2:00 minutes at a rate of 4 Hz before depositing SrRuO$_3$ at a rate of 10Hz for about 10 seconds per unit cell thickness. After deposition, the sample was cooled down at a rate of 5°C/min while the oxygen pressure was increased to 50 Pa to enable post-annealing of the film, removing impurities in the sample and allowing full oxygenation [20].

Figure 4.1: Illustration of a standard PLD setup, in which a laser beam is incident on a target in a vacuum chamber, creating a plume of material that is deposited on the substrate. Image taken from [30].

4.1.3 Atomic force microscopy

To be able to investigate the quality of our film growth, it is necessary to detect the desired step flow growth of SrRuO$_3$ on our SrTiO$_3$ substrates. As the psuedocubic unit cell height is about 4 Å, imaging at the order of fractions of nanometers is required. One easy method is atomic force microscopy (AFM), a type of scanning probe microscopy that allows relatively quick high resolution imaging in ambient conditions. A major advantage of AFM above other high resolution microscopy methods, such as scanning tunneling microscopy (STM) is that as AFM works with force
principles, imaging of both conducting and insulating samples is possible, whereas STM only works with conducting samples. In our experiments the AFM tapping mode is used, where a cantilever with a tip scans the surface of the sample with a frequency similar to or slightly lower than the resonance frequency of the cantilever. Due to variation in repulsive Coulomb and attractive Van der Waals interactions as the distance between the tip and the sample is changed, the cantilever resonance frequency is changed as well. Moreover, as the tip is closer to the surface the oscillations are damped. These changes in amplitude and frequency are measured by using the deflection from a laser incident on the cantilever. This information is used both to provide feedback to the system in order to change the cantilever height as to keep the amplitude oscillation constant, and to calculate the height of the sample for all points in a 2D plane. In this way, high resolution height maps can be produced. Furthermore, imaging can be improved by adjusting the gain and the scan rate, where a higher gain increases the feedback reaction speed at the cost of more noise, while increasing the scan rate provides better quality imaging at the cost of time [31].

4.1.4 X-ray diffraction

X-ray diffraction is a method for identifying different phases of crystalline material and studying properties such as crystal orientation, crystallite size and lattice structure. Using a combination of known and observed data, film and substrate properties can be studied for our purposes. X-rays of a wavelength of usually 1.54 Å are produced by firing electrons on a cooled metal target and filtering the emitted radiation. The reason for using X-rays of these wavelengths is that they are similar to interatomic distances in order for the crystal structure to diffract the X-rays. The basic principle of X-ray diffraction is the following. As X-rays are incident on the atoms in the material oriented in a crystal structure, the X-rays get scattered by the electrons around the atom nuclei. Due to the crystalline order, at certain angles the scattered rays will constructively interfere. The necessary condition is defined by Bragg’s law,

\[ 2d \sin(\theta) = \lambda n. \]  

(4.1)
As the constructive interference will only occur when the path difference of the rays is equal to an integer amount of wavelengths, Bragg’s law connects the geometric condition with the used wavelength. Here, \( d \) is the spacing between the crystal planes, \( \theta \) is the angle of incidence, while \( \lambda \) is the wavelength and \( n \) is an integer. A schematic of the geometry is shown in Fig. 4.2.

\[ d \sin \theta = n \lambda \]

\( \text{Figure 4.2: Schematic of Bragg-diffraction.} \)

The reflected rays are collected using a detector, such as a proportional counter. The angle that is varied is usually \( 2\theta \), the angle between the transmitted beam and the diffracted beam. By measuring the intensities on the detector for different values of \( 2\theta \) while the angle of incidence is kept equal to the angle of diffraction, the different Bragg peak positions and intensities related to the crystal planes of the sample can be identified, giving information about the different phases present, as well as the lattice parameters and information on the strain in thin films. This is called a 2 theta-omega scan. The peak width and observable fringes can be related to the thickness of the SrRuO\(_3\). By indexing diffraction patterns (assigning a set of atomic planes to each peak) information about the crystal structure and orientation can be found. For example, crystals with different symmetry groups can have different absent peaks, and textured or oriented crystals will have different relative peak intensities compared to their powder form. Finally, for a specific Bragg angle, the detector can be fixed while varying the angle \( \omega \) between the incoming beam and the sample surface. This can be done to study how well a certain phase is oriented, thereby giving information on epitaxial film quality [32–34].
4.1.5 Magnetic property measurements

Magnetic properties were measured in a magnetic properties measurement system (MPMS). This system makes use of a SQUID (superconducting quantum interference device) including two Josephson junctions in a superconducting loop. Due to the magnetic flux quantisation, changes in magnetic fields can be measured with great accuracy [35]. Using these features, measurements of the total magnetic moment as a function of applied field and temperature can be done in order to give information about the magnetic properties of the sample, such as the presence of a ferromagnetic film and the Curie temperature to separate different magnetic phases.

4.1.6 Longitudinal transport measurements

Electrical transport measurements for measuring the resistance of the sample as a function of temperature were done to compare the low temperature resistivity behaviour of ultra-thin films of SrRuO$_3$ to literature values, distinguishing various film thicknesses. The measurements were done using a Cryogenic cryostat and were performed in the Hall bar geometry measuring the longitudinal resistance $R_{xx}$, i.e. the resistance along the Hall bar. A current of 10 µA was applied along the bar in a four-point measurement setup, where the voltage difference was measured at two points between the current contacts as to prevent the measurement being influenced by the contact resistance. The separation between the voltage contacts is 1.2 mm. Resistance was converted to resistivity and compared to literature values.
4.2 Hall measurements

4.2.1 Device fabrication

A common process for creating devices from thin films is photolithography in combination with etching and lift-off techniques [36]. In order to study different Hall effects, transport measurements using a Hall bar configuration are required, which is fabricated using a combination of lithography and etching techniques described below.

As our films are ultra-thin, this creates an additional risk of damaging the film while bonding to the contact pads. To prevent this, 40 nm thick gold contact pads are deposited on the SrRuO$_3$ using sputtering and lift-off techniques. First of all, the sample is cleaned by soaking it in acetone and isopropanol followed by drying it with a flow of N$_2$. The AZ 4533 photoresist is then spin coated on the sample for 1 minute at 8000 rpm to provide an uniform thickness over all of the sample. The resist is then baked onto the sample by placing the sample on a hot plate for 1 minute at 100°C to densify the resist. Using a mask aligner, a mask with holes for six contact pads is brought into contact with the resist on the sample. For our devices the resist is then exposed to UV light for 10 seconds. The basic idea is that only the parts of the resist not covered by the mask get exposed, in order to be able to create the desired shape. After the exposure the sample is soaked in AZ 351 B developer mixed with deionised water (1:3) for 40 seconds, which dissolves the exposed photoresist and only leaves a layer of resist on the nonexposed parts. This lithography process is illustrated in Fig. 4.3. Hence, we end up with a sample covered in photoresist except for the contact pad locations. Now gold is sputtered on top of the sample using an Emitech sputter coater. Subsequently, the sample is soaked in acetone for 30 minutes as to dissolve the unexposed resist, removing with it the gold on top. This is the lift-off process. If necessary, a short period of ultrasonication is used. Consequently, gold only remains in the shape of the contact pads.

![Figure 4.3: The photolithography process.](image)
4.2 Hall measurements

In order to create the Hall bar structure, another layer of resist is spin-coated on the sample. Now photolithography is used with a mask that covers the shape of a Hall bar, including the priorly sputtered gold contacts, while the rest of the mask being transparent. After exposure and developing, resist only covers the SrRuO$_3$ in a Hall bar pattern, while the rest of the film is exposed. The exposed film is then etched away using an argon ion miller, whilst the Hall bar pattern is protected. For our purposes the parameters need to be optimised such that the SrRuO$_3$ is completely etched away, while not overly damaging the SrTiO$_3$ substrate. After the milling the residual resist is washed away with acetone, leaving the substrate with on top of it a Hall bar structure of SrRuO$_3$ while its contact pads are covered in gold. This device and its dimensions is schematically shown in Fig. 4.4 and is ready for measurement.

![Image of a Hall-bar structure after deposition of gold contacts, lift-off and etching.](image)

**Figure 4.4:** Image of a Hall-bar structure after deposition of gold contacts, lift-off and etching.

4.2.2 Hall effect transport measurements

Hall effect transport measurements where done to investigate the AHE and the THE of the ultra-thin SrRuO$_3$ films. The measurements were performed using a Cryogenic cryostat combined with a Stanford Research SR830 DSP lock-in amplifier to measure the transverse resistance $R_{xy}$ of the Hall bar. As the signals of both the AHE and THE were too weak to detect using the internal electrical setup of the cryostat, the lock-in amplifier was connected to the circuit. The lock-in output was used to set an internal AC source voltage of 150 mV that applied to a resistor of 15 kΩ, so that a current of 10 µA was applied to the sample. A magnetic
field was swept up to ± 3 T. The lock-in input was connected to two contact pads in order to measure the transverse resistance. The desired signal was calculated by the lock-in by comparing the original (reference) signal and the measured signal. Resistance was then converted to resistivity and compared to literature values.

As there is always a contribution of the longitudinal magnetoresistance present in the transverse resistance signal, this is filtered out by applying an antisymmetrisation procedure on the raw data. The positive and negative field sweep data were separated and subsequently interpolated on the same field coordinates. Using the following relations the data was antisymmetrised [23]:

\[
\rho_+^\prime = \frac{\rho_+(H) - \rho_-(-H)}{2} \tag{4.2}
\]

\[
\rho_-^\prime = \frac{\rho_-(H) - \rho_+(-H)}{2}. \tag{4.3}
\]

4.2.3 Gating experiments

For gating experiments another gold electrode was made on top of the sample next to the Hall bar at a distance of approximately 200 μm by photolithography and sputtering. A backgate was created by putting silver paste on the back of the 0.5 mm thick SrTiO₃ substrate. Voltages were applied between the topgate and the backgate and Hall measurements were performed in the same manner as described above.
Results and Discussion

5.1 SrRuO$_3$ growth

Regarding the SrRuO$_3$ growth process different improvements have been made, especially considering the growth quality. As described in Section 4.1, initially an annealing process was used to make the SrTiO$_3$ substrates TiO$_2$ terminated. However, we found that the desired quality of the termination was not as expected, resulting in the growth of trenches. An example of this trench growth is shown in Fig. 5.1(a). Here, meandering islands of SrRuO$_3$ seem to be separated by 2 nm deep trenches. As a film with 4 to 5 u.c. thickness was desired and the trench depth seems to be fairly constant, the SrRuO$_3$ only seems to be grown at the island sites. The trenches show a strong resemblance with the trenches in literature shown in Fig. 3.2(a), where the SrRuO$_3$ rate of growth is expected to behave differently at different nucleation sites due to mixed termination [20]. After switching to commercial TiO$_2$ terminated SrTiO$_3$ from Crystec an even stepsize growth was found as shown in Fig. 5.1(b), where the steps are due to the angle miscut of the substrates. These results suggest that the termination realised by the initial annealing process seems to be of inadequate quality for the successful growth of ultra-thin films, whereas commercial TiO$_2$ terminated SrTiO$_3$ substrates repeatedly seem to provide good quality growth. This can be ascribed to the quality of the termination process, which is a HF-treatment in commercial substrates, yielding a better quality surface [37], but which could not be performed in our lab.
Results and Discussion

Figure 5.1: AFM height profile of SrRuO$_3$ growth on SrTiO$_3$ substrates using (a) O$_2$-annealed substrates and (b) commercial TiO$_2$ terminated substrates.

Furthermore, after making this change, AFM imaging of showed that the growth qualities of films were very constant for a large amount of samples with varying thicknesses. For the growth of ultra-thin films, a growth rate of about 10 seconds per unit cell of SrRuO$_3$ was found.
5.2 X-ray characterisation

5.2.1 Film quality

X-ray diffraction was used in the first place to characterise the quality of the SrRuO$_3$ growth on the SrTiO$_3$ substrate. The goal here is to make sure that the thin SrRuO$_3$ film has been grown epitaxially on the substrate. Whether a film is grown epitaxially on the substrate can be seen by observing only the peaks from both materials that correspond to planes with the same orientation in a 2 theta-omega scan. An example of such a scan for a 5 u.c. SrRuO$_3$ film is shown in Fig. 5.2, where only peaks with Miller indices of type (0 0 1) are observed for both substrate and film, hence the film has been grown epitaxially. In this figure, the periodical high peaks are caused by the single crystal property of the SrTiO$_3$. On top of these substrate peaks the peaks of the film are shown with a lower intensity, seemingly broadening the peaks. Although there could be other reasons for peak broadening, the main reason is the thinness of the films.

![Figure 5.2: Example of a 2 theta-omega scan for SrRuO$_3$ grown on a SrTiO$_3$ substrate. The high peaks are the substrate peaks, while the broadening at lower intensity is due to the film thickness of SrRuO$_3$.](image)

5.2.2 Film thickness

Ideally, a REED system would be used to monitor the growth of single layers of SrRuO$_3$ \textit{in situ} during deposition. Thicker films could be measured with AFM using a step edge or by X-ray reflectivity (XRR). However, as for films of a thickness of about and below 2 nm no fringes can be observed in the XRR fitting model for SrRuO$_3$, thicknesses of our films were estimated by comparing the width of SrRuO$_3$ peaks in XRD. This was done using a
Results and Discussion

model based on the kinematic approximation of diffraction. Peak broadening is caused both by the thickness of our films and by other effects such as microstrain. As our films are so thin, we assumed that the broadening of the peaks if mainly caused by the thinness of our films, while the other effects are small compared to this effect. As there could still be contributions of the other effects to peak broadening, our model can only give a lower bound to the thicknesses. As shown in Fig. 5.3, one peak of the 2 theta-omega scan was compared with this model resulting in a thickness estimation of between 4 and 5 unit cells, probably closer to 5 when other broadening effects are present. The model was calibrated by measuring the thickness of thicker films using XRR and comparing the growth rate.

Figure 5.3: Part of a 2 theta-omega scan around the SrTiO$_3$ (0 0 2) peak. The SrRuO$_3$ contribution can be seen in the broader peak. This peak was compared for different thicknesses of the film with a model. This film seems to be about 4 - 5 unit cells.
5.3 Magnetic characterisation

The magnetic properties of the grown films were characterised in the first place to prove that there is a ferromagnetic layer present, an indication for that the grown layer is indeed SrRuO$_3$. In the second place they were characterised to investigate whether the Curie temperature of the ferromagnetic transition matches to the literature values. In an $M(T)$-measurement, the graph shown in Fig. 5.4 was obtained.

**Figure 5.4:** Out of plane magnetic moment as a function of temperature for 4 u.c. of SrRuO$_3$. The measurements are done during cooling while applying a saturation field (red curve: field cool (FC)), subsequently for warming up while applying no field (a tiny 5 mT field is applied to compensate for a residual field) (green curve: field warm (FW)), and for warming up after the system has been cooled in zero field (blue curve: zero field cool - field warm (ZFC-FW)). Three ferromagnetic transitions can be observed.

Instead of one expected ferromagnetic phase transition, three ferromagnetic phase transitions are observed at around 2 K, 80 K and 110 K. None of these transitions correspond to the literature value of the Curie temperature of about 150 K for epitaxial films, but for ultra-thin films $T_C$ is found to decrease due to the itinerant origin of ferromagnetism in SrRuO$_3$ [20]. For 4 u.c. of SrRuO$_3$ $T_C$ was expected to be around 110 K and later found around 90K in a bilayer structure combined with SrIrO$_3$ (whereas $T_C \approx 120$ K for 5 u.c.) [7, 20]. These values match fairly well the phase transition at 110 K and thus could be explained by the ferromag-
Results and Discussion

The magnetic phase transition of SrRuO$_3$. The phase transition at 80 K raises more questions, but one hypothesis is that it could be caused by another ferromagnetic phase in our samples. Another possibility regarding the fact that the magnetic moment consists of a very weak signal due to the thinness of the film is that it could be caused by particulates on the substrate, for example caused by imperfect deposition or cutting the substrates by the supplier. The last phase transition around 2 K is thought to be due to the SrTiO$_3$ substrate, in which impurities or interface effects have shown to produce some ferromagnetic signal at low temperatures [38].
5.4 Transport measurements

Electrical transport measurements for measuring the longitudinal resistivity $\rho_{xx}$ were performed on films with expected thicknesses of 4 and 5 unit cells. Our results in Fig. 5.5(a) have been compared to literature values in Fig. 5.5(b). One notable difference between literature and our results is that [7] used a two unit cell capping layer of SrIrO$_3$, however this is not expected to influence any longitudinal transport properties. Comparing our data with the literature values, it can be concluded that there are clear similarities between both the resistivity behaviour and values for both thicknesses. Our data was primarily focussed on the low temperature regime as to find clues for the differences in expected behaviour for 4 and 5 unit cell thicknesses. According to [7] the higher resistivity of the 4 unit cells film and the hooked turn below 50 K are due to the localisation of electrons for lower thicknesses, forming a nearly insulating state at 4 unit cells thickness. This is in accordance with earlier findings that films of 3 unit cells thickness and thinner are found to be insulating [20].

![Figure 5.5: Longitudinal resistivity $\rho_{xx}$ of (a) measurements of 4 and 5 u.c. SrRuO$_3$ compared with (b) literature values taken from [7]. Here, $m$ represents the number of unit cells.](image)

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5.5 Hall measurements

Hall measurements for a film with 5 u.c. thickness show a clear contribution from the AHE and a contribution that could either by ascribed to the THE or to the AHE in the two AHE phases model [9]. The antisymmetrised data is shown in Fig. 5.6, where the OHE is substracted as will be done in all figures henceforth. The hysteresis behaviour can be ascribed to the AHE, while the peaks near the coercive field can be ascribed to either the THE or two phases of the AHE. For practical purposes, we will just refer to them as THE peaks in this section. The peaks are visible up to 80 K and the AHE up to 100K, while becoming more visible over higher field ranges with decreasing temperature. As the Curie temperature of these films was found to be about 100 K, it makes sense that both effects appear together below this temperature because both effects can be described as a result of ferromagnetic behaviour [8]. Between 100 K and 80 K the AHE switches sign and as temperature decreases the AHE becomes both larger and grows in field range as the coercive field increases. The THE peaks arise and grow bigger between 80 K and 60 K and then stays fairly constant in resistivity values at lower temperatures. However, at these temperatures the peaks clearly stay present over larger field ranges.

Figure 5.6: Hall resistivity as a function of applied magnetic field. The data has been antisymmetrised to correct for magnetoresistance contributions and has been corrected for the OHE. The magnetic field sweep direction is shown by the red (negative to positive field) and blue (positive to negative field) colours.
Comparing these results to the literature results shown in Fig. 3.3 in Section 3.3, the data is consistent with previous results in literature for the trend in AHE and THE peak effects. In our data the magnetic fields where the AHE and THE peaks appear are found to be very similar for the different temperatures [8]. An important difference however, is the value of the Hall resistivity for the size of the AHE, which is around a factor of 5 smaller than found in the studies of [8] and [7]. Also, the maximal observed THE peaks are a factor 10 smaller than [8] and a factor of 5 smaller than [7], the latest being the study in which the capping layer was used. The origin of this difference remains unclear.

The peaks in our data and the data of [8] seem to be stable over a larger field range than in [7] for a given temperature. If the THE peaks are the effect of skyrmions this is an indication for a greater robustness of the skyrmion phase compared to skyrmion phases in other materials [3, 21]. From this data, a temperature-field phase diagram can be made, as shown in Fig. 5.7. The shape of this phase diagram where the THE phase is clearly visible as the yellow crescent in the corner and is found to be similar to the diagram found by [8]. Moreover, this phase seems larger in fields than the capping layer study or skyrmion phases in other materials [3, 7, 21].

Therefore, comparing our data with these studies, if the THE can be ascribed to skyrmions it seems that a capping layer is unnecessary and even generates a less robust skyrmion phase.

Figure 5.7: Phase diagram in the HT-plane. A ferromagnetic phase, a THE peak phase and saturated phase are shown in the figure. The colors represent the values for the Hall resistivity values.
If we compare this data to the alternative explanation of the AHE and THE as described in Section 3.4 [9], the idea of two phases of the AHE could be explained by two interfaces of the SrRuO$_3$, one with SrTiO$_3$ and one with vacuum, in sum yielding an asymmetric effect. Although in both our data and the study by [8] the THE peaks don’t disappear at low temperatures where there is no switch in the AHE sign in contradiction by what was expected by [9]. However, it still seems possible that the effect we have observed could be constructed by adding two different contributions. To make this intuitive, we fitted the data for two different AHE contributions, as shown in Fig. 5.8.

\[ \text{Figure 5.8: Hall resistivity as a function of magnetic field for various temperatures fitted for the two AHE contributions model. On the left: the data (blue) and the fit (black). On the right: two separate AHE contributions that make up the black fit on the left when summed.} \]

Especially for lower temperatures the data can be explained quite nicely with two AHE contributions continuously changing with temperature. At around 80 K both contributions seem to arise, a positive one (green) and a negative one (red). Both contributions grow in field range as temperature decreases. This is further illustrated in Fig. 5.9, where the coercive field is shown as a function of temperature.
5.5 Hall measurements

Figure 5.9: Coercive field as a function of temperature for both hypothetical AHE contributions together giving rise to a total AHE with additional peaks. Both contributions seem to arise around 90 K.

The size in resistivity of the positive contribution stays fairly constant while the negative clearly grows in size starting from 80 K. Both contributions to the saturation resistivity are shown in Fig. 5.10.

Figure 5.10: Saturation Hall resistivity as a function of temperature for both hypothetical AHE contributions together giving rise to a total AHE with additional peaks. A transition in the red curve can be seen.
Thus we can see that even at temperatures where there is no AHE sign switching and the THE peaks are broad, they can still be modeled by two AHE contributions.

On the other hand, in the study that proposed this model it was argued that in symmetric stacks, such as SrTiO$_3$/SrRuO$_3$/SrTiO$_3$ as well as SrIrO$_3$/SrRuO$_3$/SrIrO$_3$ the THE peaks are expected to be enhanced if they are caused by the presence of a skyrmion lattice [9]. As this was in contradiction to their findings, it was treated as another argument in favour of this model. However, having two similar interfaces at both sides at the SrRuO$_3$ would yield two opposite DMI vectors, canceling out each other, so that no skyrmion lattice will be created. This means that in both models a symmetric structure would yield no additional peak in the hall measurement, so this argument cannot be used to distinguish between the two models.
5.6 Gating measurements

In earlier work on probing the AHE and THE by an electric field [28], it was argued that tuning the electric field was only possible in multilayers with 2 u.c. SrIrO$_3$ between the SrTiO$_3$ substrate and the SrRuO$_3$ film. As no THE peaks were observed in this study for a single 5 u.c. SrRuO$_3$ film, contrary to the work of [8] and our data, this raised questions about these findings. We therefore repeated the gating experiment as we expected that it might be possible to probe the THE peak by an electric field in a SrRuO$_3$ sample without capping layer. The Hall resistivity as a function of applied field and gate voltage is shown in Fig. 5.11.

**Figure 5.11:** Hall resistivity at 2.6 K as function of applied field for various gating voltages ranging from -200 V to 200 V.

The study of [28] showed that the AHE could be increased by 35 nΩ · cm when applying a negative voltage of -180 V and decreased by 50 nΩ · cm by applying a positive voltage of 200 V with respect to the AHE signal at 0 V, all at a temperature of 2 K. This relatively large shift was not observed in our data, where only a very small shift maximally varying 10 nΩ · cm was observed, as shown in Fig. 5.12(a). Compared to the trend in literature values, there seems to be not much of a shift in our data. One possibility is that a capping layer is necessary to be able to probe the AHE by an electrical field as proposed by [28], while the minor shift we are seeing is due to some other effect. In this view, the electric field primarily affects the SOC, resulting in a deformation of the band structure in k-space and therefore a switch in the AHE. A second possibility could be that the quality of gating was not as desired, resulting in a weaker electric field af-
fecting the film. Another possibility is that some not yet well-understood process is going on in which the electrical field influences the film. There was no correlation found between the signal and temperature instability.

The same study as mentioned above showed a shift of decreasing the THE peak by about 15 nΩ·cm when applying a negative voltage of -180 V and increasing the THE peak by more or less the same amount when applying a positive voltage of 200 V with respect to the AHE signal at 0 V, all at a temperature of 2 K [28]. This is nearly 18 % of the size of the THE at 0 V. In our data, shown in Fig. 5.12(b) a variation in the same magnitude was found making up to a similar percentage of the total THE peak at 0 V, but there seems to be no similar relationship as the shift seems to decrease somewhat from -180 V to 0 V, after which it increases again. After all, it seems that the THE peaks are affected by the field as there is some kind relationship observed. If the THE is indeed due to a skyrmion lattice, tuning the THE with an electrical field means that the size of skyrmions could be probed in this manner [28]. The absolute sizes of the THE peaks cannot be compared, as different offsets resulting from different analysing methods have been used.

![Figure 5.12: (a) The change of the AHE with respect to the AHE at zero voltage around zero field, compared to the shift in literature. (b) The change of the maximum value of the THE with respect to the maximum THE at zero voltage, compared to extrapolated data from literature.](image)

In theory it still remains unclear how the SOC is precisely affected by the electrical field, but it is suggested to have to do with the interface potential gradient due to electrical control of Rashba-type band splitting [28, 39]. However, different effects resulting from SOC seem to be involved, as we found that in ultra-thin SrRuO$_3$ films the THE peak could be probed convincingly, while the AHE is only shifted by a small amount. More research is needed to gain a precise understanding of the processes causing these effects.
Chapter 6

Conclusion

First of all, we have succeeded in growing good quality ultra-thin films of SrRuO$_3$ epitaxially on SrTiO$_3$. The thicknesses of these films were estimated by analysing the peaks using XRD with a calibrated model and verified by comparing R(T) data with literature values.

Secondly, for a 5 u.c. SrRuO$_3$ Hall bar the AHE and additional peaks were observed in Hall resistivity measurements as a function of field for a variety of temperatures. It was verified that no capping layer to enhance SOC is necessary to observe the extra effect. The phase of this extra effect in the $HT$-plane corresponds to literature value, whereas the size of resistivity of the total effect is found to be smaller. The additional peaks could either be explained by a THE, where a skyrmion phase gives rise to an extra term in the Hall effect, or due to two phases of the AHE caused by two asymmetric interfaces of SrRuO$_3$ with SrTiO$_3$ and with vacuum. In the case of a THE by skyrmions, ultra-thin SrRuO$_3$ is a good material for creating robust skyrmion lattices offering many opportunities for further research.

Finally, gating experiments have shown that the AHE is barely affected by probing with an electric field in 5 u.c. films without a capping layer, in accordance with literature. We have shown that the additional peaks that were observed could be shifted with an electric field, whereas the exact behaviour is not yet fully understood. If the effect is a THE due to a skyrmion lattice, this means that the skyrmion size could be affected, offering an easy method for the tuning of skyrmions.
Chapter 7

Outlook

As a continuation of this work, Hall effects in films of more thicknesses of SrRuO$_3$ could be measured and compared with existing literature values. This includes both films of 4 u.c. thickness that are expected to show the THE peaks and control measurement of larger thicknesses to see if the THE peaks disappear for higher thicknesses as expected.

In order to settle the debate about the presence of skyrmions in SrRuO$_3$ one would ideally choose a direct approach such as imaging skyrmions. The feasibility of Lorentz tunneling electron microscopy (LTEM) has been investigated. Optimistic estimations using a thickness of 2 nm and skyrmion size of 20 nm [7], together with a volume magnetisation of 1.4 $\mu_B$ per Ru, assuming a hexagonal lattice yields a contrast of 0.390% using the contrast formula by [40]. In literature 1-3 % is usually considered the minimum detectable contrast, therefore it would be very hard to prove the presence of skyrmions using LTEM. Another method is magnetic force microscopy (MFM), but this is generally limited to a resolution of about 30 nm, making skyrmion observation rather difficult [7].

When the exact dynamics giving rise to the THE peaks are found to be best understandable by the skyrmion model, new gating experiments could be performed to investigate more thoroughly skyrmion probing by an electric field, which could be interesting for the future role of skyrmions in spintronics. In the skyrmion scenario, it would also be useful to see how a superconductor will couple to the skyrmion lattice and investigate the interesting physics this could bring about, such as flux pinning. Furthermore, it was predicted that magnetic skyrmions could host Majorana bound states when placed in proximity with an s-wave superconductor, showing that skyrmions could play a role in topological quantum computing.
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Supplementary I: raw data Hall measurements

The raw temperature dependence data for R(H) measurements on a 5 u.c. SrRuO$_3$ Hall bar is shown in Fig. 7.1 for a current of 10 $\mu$A and for a current of -5.33 $\mu$A in Fig. 7.2.
Figure 7.1: Raw temperature dependence data for R(H) measurements on a 5 u.c. SrRuO$_3$ Hall bar. A current of 10 µA was applied.
Figure 7.2: Raw temperature dependence data for R(H) measurements on a 5 u.c. SrRuO$_3$ Hall bar. A current of -5.33 $\mu$A was applied.
Bibliography


