Chapter 6

Anisotropy and Current Control of Magnetisation in SrRuO\textsubscript{3}/SrTiO\textsubscript{3}

Abstract

Spintronics-based nonvolatile components in neuromorphic circuits offer the possibility of realising novel functionalities at low power. Current-controlled electrical switching of magnetisation is actively researched in this context. Complex oxide heterostructures with perpendicular magnetic anisotropy (PMA), consisting of SrRuO\textsubscript{3} (SRO) grown on SrTiO\textsubscript{3} (STO) are strong material contenders. Utilising the crystal orientation, magnetic anisotropy in such simple heterostructures can be tuned to either exhibit a perfect or slightly tilted PMA. Here, we investigate current-induced magnetisation modulation in such tailored ferromagnetic layers with a material with strong spin-orbit coupling (Pt), exploiting the spin Hall effect. We find significant differences in the magnetic anisotropy between the SRO/STO heterostructures, as manifested in the first and second harmonic magnetoresistance measurements. Current-induced magnetisation switching can be realised with spin-orbit torques, but for systems with perfect PMA this switching is probabilistic as a result of the high symmetry. Slight tilting of the PMA can break this symmetry and allow the realisation of deterministic switching. Control over the magnetic anisotropy of our heterostructures, therefore, provides control over the manner of switching. Based on our findings, we propose a three-terminal spintronic memristor, with a magnetic tunnel junction design, that shows several resistive states controlled by electric charge. Non-volatile states can be written through SOT by applying an in-plane current, and read out as a tunnel current by applying a small out-of-plane current. Depending on the anisotropy of the SRO layer, the writing mechanism is either deterministic or probabilistic allowing for different functionalities to emerge. We envisage that the probabilistic MTJs could be used as synapses while the deterministic devices can emulate neurons.

This chapter is based on work published in:
6.1 Introduction

Current information processing technologies make use of the von Neumann architecture. While this approach is suitable for performing simple iterative operations, it is not as well equipped for more complex tasks such as recognition and prediction. The brain, however, can tackle these tasks with considerable ease and for this reason, progressively more research is being conducted into brain-like computation. The field of artificial intelligence, for example, has seen great success in developing software that emulate the functioning of the brain. However, these software still run on von Neumann hardware, making this approach the subject of several limitations, such as high power usage. Development of novel hardware that can directly mimic brain-like neural networks is therefore of significant interest. The main components of the brain are neurons, which process information, and synapses, which learn and remember by adjusting their connection strength. In particular, neurons are often regarded as stochastic spiking elements that transmit digital signals across analogue synapses, whose conductance can take on a range of values\(^2,3\).

Spintronics, where information can be stored in a non-volatile way using magnetic states, offers an attractive platform for emulating these brain components in solid-state devices. Here, we focus on systems with perpendicular magnetic anisotropy (PMA), where the magnetisation can be either parallel or antiparallel (pointing ‘up’ or ‘down’) to the film normal, making them suitable candidates for binary memory elements. PMA systems have various advantages over their in-plane easy axis counterparts, including enhanced thermal stability of the magnetic ordering, higher data storage density, improved scalability and more facile writing of the states\(^4,5\). Realising analogue states with the aid of magnetic domains\(^6\) is also possible. The presence of the domains allows obtaining magnetic states where some domains point up and some point down so that the overall magnetisation assumes a magnitude lower than the saturation value. The possibility of achieving analogue states, largely absent in standard electronics, is particularly useful for emulating brain-like behaviour which is believed to make use of a combination of analogue and digital signals\(^7\).

In order to write the magnetic state, it is important to be able to switch the magnetisation orientation. When the ferromagnet is placed adjacent to a heavy metal layer, its magnetisation orientation can be switched with an in-plane current owing to the presence of spin-orbit torques induced by the spin Hall effect and Rashba fields\(^8-10\). SOT switching has also been realised in a single-crystalline ferromagnetic layer with intrinsic bulk inversion asymmetry, such as GaMnAs\(^11\). Here the Dresselhaus- and Rashba-like fields give rise to current-induced torque. A potential downside of the latter for memristive applications is that the single crystalline nature of the ferromagnet does not allow for intermediate magnetisation states (between up

and down) to be realised.

Systems with perfect PMA are symmetric with respect to these torques, rendering the magnetisation switching probabilistic. Deterministic switching, on the other hand, can be realised by breaking the symmetry in the system. The simplest way of breaking this symmetry is via the application of a small in-plane field, but this is not an ideal solution from the viewpoint of technological applications. Other approaches include introducing asymmetry through exchange bias\cite{12,13}, device geometries with lateral structural asymmetry inducing an anisotropy gradient\cite{14}, or through a precise control of the magnetocrystalline anisotropy of the ferromagnet\cite{15,17}.

Transition metal complex oxides have the unique potential for controlling their magnetocrystalline anisotropy via relatively straightforward means due to the intimate relationship between their crystal structure and magnetic properties. In this chapter we illustrate this using SrRuO$_3$ (SRO).

6.2 Magnetic Anisotropy

Magnetic anisotropy refers to how direction affects the magnetic property of a material. It describes how a material might be easier or more difficult to magnetise depending on the direction of the applied magnetic field w.r.t. the material. There can be several contributions to this phenomenon. Shape anisotropy; this is usually observed in materials that are not spherical symmetric as the demagnetising field will not be the same in all directions. Magnetoelastic anisotropy; this is magnetic anisotropy caused by tension or force on a material. Exchange anisotropy; the magnetic interaction between adjacent magnetic moments. And magnetocrystalline anisotropy; this is an intrinsic contribution to the anisotropy and refers to the energy to magnetise a material being different depending along which crystallographic axis the field is applied. The primary source of this type of anisotropy is spin-orbit interaction.

6.2.1 Perpendicular Magnetic Anisotropy

For technologically viable magnetic devices where a high packing density is required, it is desirable to utilise ferromagnetic layers with perpendicular magnetic anisotropy (PMA); i.e. layers where the magnetic easy axis lies along the normal direction. This allows the magnetic moments to align side-by-side (versus the head-to-tail alignment present in layers with an in-plane easy axis), resulting in higher thermal and magnetic stability, lower power consumption as well as an increased data storage density.

In thin films, magnetisation typically prefers to lie in the plane of the film due to
6.2. Magnetic Anisotropy

Figure 6.1: SrRuO$_3$ crystal structure showing the orthorhombic unit cell (superscript $o$) and the pseudocubic cell in thick solid lines (superscript $ps$). Based on [18].

the demagnetising energy cost associated with the out-of-plane orientation. Realising a system with PMA, therefore, requires compensation of this energy cost. In ferromagnetic/heavy metal multilayers, for example, the translational invariance lost in the direction of the film normal gives rise to an additional interfacial anisotropy. This can overcome the demagnetising energy and yield PMA in the ferromagnet. However, this is an interfacial effect and hence limited to ultrathin ferromagnetic films, which can be challenging to fabricate[19–21]. Crystalline alloys of Fe and Co, such as FePt, FePd and CoPt have also been investigated as PMA materials. In these materials the L1$_0$ phase has been found to have a large uniaxial magnetic anisotropy resulting in an out-of-plane magnetisation[22–24]. A drawback of these materials, however, is that their magnetisation dynamics suffer from strong damping and hence significant energy dissipation. This aspect renders them less suitable for technological applications[5, 24, 25]. Other material systems exhibiting PMA include the ferromagnetic Heusler alloys[26, 27] with uniaxial anisotropy favouring out-of-plane magnetisation, and amorphous rare-earth transition metal alloys[28–30], where the combination of strong exchange interactions and strong magnetic anisotropy allows
tailoring of the latter. A material that exhibits strong magnetocrystalline anisotropy both in bulk and thin films is SrRuO$_3$; hence, depending on the orientation of the crystal direction in thin films, it can be used to realise PMA.

### 6.3 SrRuO$_3$

SrO belongs to the class of Ruddlesden-Popper ruthenate phases $Sr_{n+1}Ru_nO_{3n+1}$, where specifically $n = \infty$. At room temperature, the Sr-O bonds are significantly shorter than the Ru-O bonds causing the RuO$_6$ octahedra to rotate. As a result, SrO has an orthorhombic symmetry with lattice parameters $a=5.5670$ Å, $b=5.5304$ Å, and $c=7.8446$ Å; this structure can be approximated as pseudocubic with a lattice constant of 3.93 Å. At higher temperatures the octahedral rotations are reduced, causing transitions to higher crystal symmetries. Specifically, around 550$^\circ$C SrO becomes tetragonal while around 680$^\circ$C a transition to a perfect cubic perovskite structure occurs.

![Diagram](image)

**Figure 6.2:** The orbital energy of (b) a free Ru$^{4+}$ ion and (a)&(c) in an octahedral crystal field. The crystal field lifts the degeneracy of the 4d-orbitals. Depending on the energy gap between the energy levels, the electrons can either be in a (a) high or (c) low spin state.

In the high-temperature cubic phase, there is a spherically symmetric electric field around each Ru ions, originating from repulsion between the six coordinating oxygen ions. This results in all five 4d-orbitals being degenerate as shown in Fig. 6.2(b); this degeneracy is lifted when the oxygen ions adopt an octahedral configuration at lower temperatures. The octahedral field results in three degenerate lower energy levels with $t_{2g}$ symmetry and doubly degenerate higher energy levels with $e_g$ character (Fig. 6.2(a)+(c)). Ru has four valence electrons giving rise to a ground...
Figure 6.3: Schematic representation of the orientation of $\text{SrRuO}_3$ thin films on (a)-(c) $\text{SrTiO}_3$ (001) and (d)-(f) $\text{SrTiO}_3$ (110) substrates. (b), (c), (e) and (f) show 2D cuts along different crystal planes where the top and bottom coordinate systems refer to orthorhombic SrRuO$_3$ and (pseudo)cubic SrTiO$_3$/SrRuO$_3$ notations respectively. Inspired by [34].

state of $t_{2g}^{3}t_{2g}^{1}$ following Hund’s rules; the partial filling of the minority spin states compared to the full occupation of the majority $t_{2g}$ spin band makes this state ferromagnetic [35,36]. This is unique to SRO as the extended orbitals of 4$d$ systems give rise to a wider band, resulting in a reduction of the densities of states at the Fermi level. The increased density of states at the Fermi level in SRO is the result of the strong hybridisation of the 4$d$ and 2$p$ states and makes it the only itinerant ferromagnetic 4$d$ transition-metal oxide [37–39]. Usually, the crystal field-induced splitting will be relatively large due to the diffuse nature of the 4$d$ orbitals, making it energetically favourable to place all four electrons in the lower-lying $t_{2g}$ orbitals. This gives rise to a low-spin configuration, with two unpaired electrons and a spin angular momentum of $S = 1$ resulting in a magnetic moment of $2\mu_B$ (Fig. 6.2(c). It has, however, been observed that the application of sufficient compressive epitaxial strain can cause distortion of the Ru octahedral environment resulting in one of the electrons favouring the $e_g$ band. In this case, the Ru can be in a high-spin state where

$S = 2$ and $\mu_{Ru} = 4\mu_B$ (Fig. 6.2(a)[40].

The average spin polarisation of the conduction electrons at the Fermi surface is in the opposite direction to the magnetisation, resulting in SRO being classified as a minority-band ferromagnet. This has been confirmed from band structure calculations, which predict small and negative spin polarisations, ranging from -9%[39,41] to -20%[38] and experimentally in magnetic tunnel junctions (P=-9% - -9.5%)[42,43].

6.3.1 Magnetocrystalline Anisotropy

The magnetocrystalline anisotropy of bulk SRO is complicated and may depend on the crystal symmetry and temperature[36,44,45]. In general, SRO is well-known for its strong magnetocrystalline anisotropy stemming from the strong spin-orbit coupling of the heavy Ru ions. The magnetic easy axis of SRO has been observed to lie close to the crystallographic orthorhombic $b$-axis ([010]$_o$), which corresponds to the [110]$_{pc}$ direction[18,46]. Below $T_c$, a gradual orientation transition occurs where the $b$-axis moves closer to the normal direction at a rate of $\sim 0.1^\circ/K$, reaching $\sim 30^\circ$ at low temperature. The uniaxial anisotropy energy as a function of the angle between the magnetisation and easy axis, $\theta$, is given by:

$$E_{anisotropy} = K \sin^2 \theta,$$

where $K$ is the anisotropy constant with a value of about $1.2 \times 10^7$ ergs/cm$^3$ and a corresponding anisotropy field ($\frac{2K}{M}$) of $\sim 12$ T[36,47].

For this reason, the orientation of the unit cell will effectively determine the easy axis direction. Epitaxial thin films of SRO can be grown on SrTiO$_3$ (STO) substrates, as the latter has a perovskite structure with a closely matching lattice constant. Interestingly, the SRO unit cell orientation tends to adopt that of the STO substrate to minimise the in-plane strain[48]. This provides us with an elegant tool for controlling the magnetocrystalline anisotropy of the SRO film without the requirement of ultrathin films, additional layers, or complex layer structures and device geometries.

Here, we have utilised STO substrates with two different orientations, (001) and (110), where the Miller indices refer to the crystal planes parallel to the surface. As discussed above, the SRO films grown on these substrates are expected to adopt (001) and (110) orientations, respectively (illustrated in Fig. 6.3). Referring to the relationship between the crystal structure and the magnetocrystalline anisotropy, we would anticipate that the magnetic easy axis to lie along the film normal for the (110) film, while in the (001) film it is predicted to have an in-plane component. Due to the absence (presence) of this in-plane component, we expect the former (latter) to exhibit probabilistic (deterministic) switching. In order to enable current-induced switching of the SRO layer, a layer of strongly spin-orbit coupled Pt has been included in our device structures to serve as a source of spin-orbit torque. The heterostructures with
different magnetocrystalline anisotropies and switching properties investigated in this work can potentially serve as spintronic memristive materials. By incorporating them in a three-terminal magnetic tunnel junction design, deterministic switching devices can emulate a spin-synapse whereas probabilistic switching devices can function as neurons.

6.4 Materials and Methods

STO substrates with (001) and (110) orientations with small miscut angles were treated with buffered hydrofluoric acid and annealed in oxygen. We deposited SRO films on the STO substrates by pulsed laser deposition (PLD). A KrF excimer laser beam ($\lambda = 248$ nm) was focused onto an SRO target at a repetition rate of 1 Hz, with a laser fluence of $1.5 \text{ Jcm}^{-2}$. The films were deposited at $600 \, ^\circ\text{C}$ with an oxygen pressure of $0.13 \, \text{mbar}$. After 1200 laser pulses, we cooled the films down to room temperature under an oxygen pressure of $100 \, \text{mbar}$\(^{[49]}\). Throughout this chapter, we will refer to the SRO/STO (001) samples as SRO\(_{001}\) and to the SRO/STO (110) samples as SRO\(_{110}\).

We performed structural and magnetic characterisation measurements on the films before fabricating devices. To determine the unit cell orientation of the SRO films on the different substrates we used different X-ray diffraction (XRD) techniques at room temperature. X-ray reflectivity measurements were used to estimate the thicknesses of the SRO layers. $2\theta$-$\theta$ scans were performed to determine the out-of-plane lattice constant. We also conducted reciprocal space mapping (RSM) to infer information about the in-plane lattice parameters of the SRO films. Finally, azimuthal scans, where a diffraction peak appears whenever a reciprocal lattice point is crossed, were carried out and information about the film alignment relative to that of the substrate was inferred from their relative peak positions. For identical alignments, identical peak positions are expected.

Temperature and field-dependent magnetic measurements were performed using a superconducting quantum interference device (SQUID) magnetometer (Quantum Design, MPMS). The different field orientations adopted were $0^\circ$ (in the plane of the film), $90^\circ$ (along the film normal) or at $\sim 45^\circ$ (diagonal to the film normal).

Post structural and magnetic characterisation, we sputtered a Pt layer on the SRO films for current-induced switching studies. The Pt/SRO bilayers were etched into Hall bars oriented along different crystallographic axes using UV lithography and ion beam etching techniques. Schematic diagrams of the Hall bars are shown in Fig. 6.11 and Fig. 6.12 where the SRO and Pt layers are indicated in red and blue respectively. Electrical measurements were done by applying an alternating current (AC) along the $x$-axis of the Hall bar. The first and second harmonic voltage responses
Anisotropy and Current Control of Magnetisation in SrRuO$_3$/SrTiO$_3$ were measured simultaneously in both the longitudinal (along the x-axis parallel to the current) and transverse (along the y-axis perpendicular to the current) direction. It should be noted that $xz$ and $yz$ scans were conducted on Hall bars of different, perpendicular orientations.

### 6.5 Characterisation of SrRuO$_3$ Films

#### 6.5.1 PLD Growth

![RHEED for SRO/STO (001). The diffraction pattern (a) before and (b) after deposition. (c) Depicts the intensity over time of the three diffraction spots marked in (a). The arrows indicate the start and end of the deposition process.](image)

Thin films of STO were grown on STO (001) and STO (110) substrates using PLD. In situ reflection high energy electron diffraction (RHEED) was used to monitor the
growth, as shown in Figs 6.4 and 6.5. Prior to SRO deposition, the RHEED patterns of the bare STO substrates are shown in Figs 6.4(a) and 6.5(a). Both show three diffraction spots lying on the zeroth order Laue circle, indicating a smooth and crystalline surface. The patterns, however, deviate from each other due to the different orientations of the STO and consequently their reciprocal lattices. At the start of the deposition, both samples show a small number of intensity oscillations which quickly die out, depicted in Figs 6.4(c) and 6.5(c). The loss of oscillations for the two samples has different origins, evident from the diffraction patterns. For SRO grown in STO (001), the RHEED pattern after growth shows no significant changes in the spot positions (Fig. 6.4(b)). This indicates that during deposition the growth mode changes from the initial layer-by-layer growth to a step-flow mode, commonly observed for SRO/STO. For the STO (110) sample, on the other hand, during deposition the RHEED pattern changes from having diffraction spots lying on the Laue circle towards a regular grid of spots, displayed in Fig. 6.5(b). This is characteristic of 3D island growth, resulting from the electron beam passing through the islands rather than reflecting off the film surface. Instead of a reflection pattern, the pattern on the screen results from the transmission. The reciprocal lattice of the islands resembles that of a bulk crystal resulting in a transmission pattern composed of an array of reciprocal points.

Post-deposition, atomic force microscopy (AFM) was used to probe the surface topography. For SRO(001), trenches can be seen on the surface, which occurs when the film growth is slower in some regions than on the rest of the film. This selective growth is commonly observed when the starting surface has mixed termination, due the faster growth rate of SRO on TiO\textsubscript{2} terminated STO compared to SrO terminated STO\textsuperscript{36}. The effect of the mixed termination is expected to diminish when the film thickness increases and eventually the areas surrounding the trenches coalesce. The RMS roughness between the trenches is found to be around 0.1 nm. The AFM scan for SRO(110) confirms the island growth; the surface consists of small protrusions, and the RMS roughness of the film is around 2 nm. This is considerably higher than that of the film grown on STO (001). The island growth is likely due to the starting termination of the substrate being non-ideal, resulting in poorly defined terraces and discontinuous step edges. We utilised the same chemical and thermal annealing treatment for both samples. While this is a well-established surface termination protocol for STO (001), this is not the case for STO (110).

### 6.5.2 XRD

We performed structural characterisation measurements on the films before fabricating devices. To determine the unit cell orientation of the SRO films on the different substrates we used different X-ray diffraction (XRD) techniques at room tempera-

Figure 6.5: RHEED for SRO/STO (110). The diffraction pattern (a) before and (b) after deposition. (c) Depicts the intensity over time of the three diffraction spots marked in (a). The arrows indicate the start and end of the deposition process.

X-ray reflectivity measurements were used to estimate the thicknesses of the SRO layers. 2θ-θ scans were performed to determine the out-of-plane lattice constant. We also conducted reciprocal space mapping (RSM) to infer information about the in-plane lattice parameters of the SRO films. Finally, azimuthal scans, where a diffraction peak appears whenever a reciprocal lattice point is crossed, were carried out and information about the film alignment relative to that of the substrate was inferred from their relative peak positions. For identical alignments, identical peak positions are expected.

The 2θ-θ scans (Fig. 6.7) confirm the (001) orientations of both the STO substrate and SRO film. We calculated the SRO out-of-plane lattice constant to be 3.968...
Figure 6.6: Atomic force microscopy (AFM) images of the SrTiO$_3$ (001) substrate (a) after chemical termination and annealing and (c) after SrRuO$_3$ deposition. AFM images of STO (110) (b) after termination and annealing and (d) after SRO deposition.

Å, revealing a tensile strain of 0.97%. Similarly, for the sample grown on STO (110) substrate (Fig. 6.7(b)), the $2\theta$-$\theta$ diffraction peaks correspond to the 110 crystal planes, suggesting that both STO and SRO have (110) orientation. For SRO, the spacing between (110) planes was found to be 2.788 Å and slightly deviates from the bulk value of 2.779 Å, suggesting a small tensile strain.

The Kiessig fringes of SRO$_{001}$ (Fig. 6.7(c)) at low angles could be fit using the recursive Parratt formalism giving a thickness estimate of 10.7 nm. For SRO$_{110}$ (Fig. 6.7(d)) no good fit could be obtained, which is consistent with the larger surface roughness of 2 nm for SRO$_{110}$ compared to the 0.1 nm roughness of the SRO$_{001}$ surface (as seen in AFM images shown in Fig. 6.6). The thickness of SRO$_{110}$ was
estimates the step height between the film and substrate using AFM.

The 3D construction of the RSM (Fig. 6.8(a) and (b)) for both samples shows a vertical alignment of the STO and SRO peaks, indicating that the films are fully strained in-plane to match the substrate lattice constant. As a result, both films experience an in-plane compressive strain of 0.64%.

For SRO_{001}, azimuthal scans were conducted around the (001), (101) (Fig. 6.8(b)), and (111) lattice planes. In most cases, the film (top panel) and substrate (bottom panel) peaks are seen to coincide closely. The only discrepancy is for the (001) planes (Appendix Fig. 6.5), where the corresponding peaks are shifted between 6 and 10°. This might be related to the strain the SRO film experiences in the [001] direction. The matching peak positions indicate that the pseudocubic unit cell of SRO has the same orientation as the STO cubic unit cell. This type of alignment is favourable as it minimises in-plane strain [48]. Similarly, the azimuthal scans for SRO_{110} (Fig. 6.8(d))
show coinciding substrate and film peaks indicating that again the alignment of the film unit cell mimics that of the substrate.

6.5.3 Magnetic Properties

Temperature and field-dependent magnetic measurements were performed using a superconducting quantum interference device (SQUID) magnetometer (Quantum Design, MPMS). The different field orientations adopted were 0° (in the plane of the film), 90° (along the film normal) or at ∼45° (diagonal to the film normal).

To probe the magnetic anisotropy of the samples, magnetic measurements were

Figure 6.9: Magnetic anisotropy of SRO/STO. Magnetic field vs. magnetisation measurements at 10 K taken with the field at different angles with respect to in-plane for (a) SRO$_{001}$ and (b) SRO$_{110}$. (c) and (d) Summarises the relative alignment between the unit cell of the substrate and the pseudocubic unit cell of the SRO film for SRO$_{001}$ and SRO$_{110}$ respectively. The figures also indicate the approximate direction of the magnetic easy axis.

done with the magnetic field directed along different crystallographic axes. The magnetisation was measured as a function of the field strength with the field along the film normal, at 45° to the film normal, and parallel to the film surface. For SRO$_{110}$, two in-plane measurements were conducted as the two primary in-plane axes ([001] and [110]) are inequivalent. Diamagnetic contributions arising from the substrate were subtracted. We conducted these measurements at 10 K, which is far below the Curie temperature (around 150 K as determined from both magnetisation-temperature and resistance-temperature measurements (Fig. 6.10)).

As seen in Fig. 6.9(a) in SRO$_{001}$, the measurements conducted with the field directed along the in-plane direction indicate that this direction is close to a hard axis. The out-of-plane hysteresis loop, on the other hand, has a large saturation magnetisation and the largest remanent magnetisation while the measurement conducted at
45° shows the largest coercive field. This indicates that the easy axis lies between 45° and 90°, meaning that it has a considerable out-of-plane component accompanied by a tilt towards the plane of the film.

For the SRO\textsubscript{110} samples, the magnetic hysteresis loops were measured with the field aligned with the surface normal, along 45° and along the two principal in-plane directions ([001] (black) and [1\overline{1}0] (green)). Fig. 6.9(b) shows the different hysteresis loops. It is clear from the results that the out-of-plane direction is the magnetic easy axis, evidenced by the largest remanent magnetisation, saturation magnetisation and coercivity. The measurement conducted along 45° shows a reduced, but still significant, remanence and coercive field. The two in-plane loops have similar shapes with no significant coercive field or remanent magnetisation, indicative of hard axes.

For both films, the high field saturation magnetic moments are different with the field applied along different directions. This is commonly observed in SrRuO\textsubscript{3} films and this effect has several possible origins. Ziese \textit{et al.} discussed that the procedure used to subtract the diamagnetic substrate background, done by taking the slope at high fields, does not take into consideration paramagnetic contributions originating from the film itself and hence can lead to different saturation values along different crystallographic directions\cite{45}. It is also possible that because the anisotropy field of SRO is higher than the maximum applied field, full saturation is not realised along the magnetic hard axes\cite{36}. A third possible explanation is that the presence of defects prevents films from reaching full saturation\cite{50}.

### 6.6 Electrical Measurements

DC and AC electrical measurements were performed after Pt deposition and fabrication of Hall bars.

#### 6.6.1 DC Measurements

To study the temperature dependence of the resistance, 100 μA was sourced through Hall bars oriented along different crystallographic directions while the sample space was increased from 5 K to 300 K. The resistance is observed to increase with temperature, which is characteristic of metallic behaviour. This is because the mean-free path, $\ell$, the average distance an electron travels between collision events, decreases as the number of thermally-induced scattering events increases. For a ferromagnetic metal, the resistivity can be expressed as $\rho = \rho_0 + \rho_{ph} + \rho_m$, the sum of a constant residual $\rho_0$, an electron-phonon scattering contribution $\rho_{ph}$ and a spin-fluctuation scattering contribution $\rho_m$. Above the Curie temperature, the spin-fluctuation part saturates. Hence, phonons are the dominant scatterers at high temperatures, and

Figure 6.10: Temperature dependence of magnetisation and resistance. Field-cooled magnetisation-temperature measurements after high field cooling for (a) SRO/STO (001) and (b) SRO (110). The magnetic field was aligned along the film normal (red) and in the plane of the film (black). (c) Resistance versus temperature measurements for differently oriented Hall bars while sourcing 100 $\mu$A.

the resistivity is expected to increase linearly with temperature'. All graphs show a change in slope around the expected $T_C$, indicating that part of the current flows through the SRO layer and not only through Pt.

*The value of $\ell$ cannot be less than the interatomic spacing, $a$, and consequently, a metallic state should saturate at a maximum resistivity when $\ell_{\text{min}} = \ell \sim a$ [51]. For SrRuO$_3$, however, no visible saturation has been observed up to 1000 K [38], leading to its classification as a bad metal.
6.6.2 AC Measurements

In ferromagnetic materials the magnetisation orientation contributes to the resistance due to the interactions between the conduction electrons and magnetic moments, giving rise to a collection of magnetoresistance (MR) effects. The MR effects are a convenient tool for understanding various material properties as it is sensitive to for instance crystalline symmetry, magnetocrystalline anisotropy, defects, domains and domain walls[45].

In order to analyse and compare these aspects of the different films, we studied MR effects by applying a 7 T magnetic field while rotating the films from in- (0°) to out-of-plane (90°). It should be noted that the magnetisation does not exactly follow the field orientation but tends towards the easy axes. This allows for further analysis and comparison of the magnetic anisotropies of the SRO/Pt bilayers on the different substrates by studying the first harmonic longitudinal and transverse resistances. The measurements we present here were performed by applying a 1.5 mA current. Further measurements, in which the current amplitude was varied are shown in Appendix Fig. E.9 and indicate the onset of discernible second harmonic signals to occur between 0.5 and 1 mA.

The longitudinal resistance (\(R\)), measured along the current (\(x\)) direction, reflects the fact that resistance depends on the angle between magnetisation and current. Known as the anisotropic magnetoresistance (AMR), this is described by Eq. 6.2 or Eq. 6.3 depending on whether the field is rotated in the \(xz\) or \(yz\) plane respectively. \(\theta\) indicates the angle between the in-plane direction parallel (\(xz\)) or perpendicular to the applied current direction (\(yz\)).

\[
\Delta R_{xz} = (R_x - R_z) \cos^2 \theta \tag{6.2}
\]
\[
\Delta R_{yz} = (R_y - R_z) \cos^2 \theta \tag{6.3}
\]

Where \(\Delta R\) represents the change in resistance induced by the AMR. In both cases, the sign of the amplitude depends on whether the resistance is higher or lower when the field is applied in-plane with respect to out-of-plane. The field orientation scans in the \(xz\)-plane are shown in Fig. 6.11(a) (SRO\(_{001}\)) and (b) (SRO\(_{110}\)). For SRO\(_{001}\) the longitudinal resistance is minimised when the current and field are aligned and maximised when the field and current are perpendicular. An interesting feature manifesting the anisotropy in the system is the differing shapes of the dips and peaks: the minima are sharp while the maxima are significantly broader. On the other hand, the field orientation scan for SRO\(_{110}\) yields behaviour that more closely resembles the expected \(\cos^2 \theta\)-dependence with similar peak shapes for both the peaks and dips. Moreover, now the maximum longitudinal resistance is observed when the field and current are parallel while the perpendicular alignment yields the minimum resis-

Figure 6.11: First harmonic angular measurements for SRO001 (left) and SRO110 (right). Data for $xz$ sweeps are shown in (a) and (b) for the longitudinal resistance and in (c) and (d) for the transverse resistance. The $yz$ scans are shown in (e) and (f) for the longitudinal resistance and in (g) and (h) for the transverse resistance. $\Delta R$ indicates the change in resistance with respect to when the field is applied out-of-plane. The angle ($\theta$) of the magnetic field ($B$) is indicated with respect to the in-plane direction. Red and black lines are used to indicate the trace (increasing angles) and retrace (decreasing angles) scans respectively.
tance. Finally, we observe a small additional sharp feature occurring whenever the current and field point in the same direction.

Scanning the field orientation in the $yz$-plane preserves the perpendicular orientation of the field with respect to the current channel. As shown in Fig. 6.11(e) (SRO$_{001}$) and (f) (SRO$_{110}$), for SRO$_{001}$, we observe flat resistance maxima when the magnetisation lies in-plane and broader dip when the field points out-of-plane. The peaks and dips are separated by sharp transitions that occur when the field is $\pm \sim 30^\circ$ away from one of the in-plane directions. For SRO$_{110}$, on the other hand, we see sharp dips when the field points in-plane and broad peaks centred around the out-of-plane direction.

The transverse resistance ($R_H$), measured perpendicular to the current ($y$) direction, results from the anomalous Hall effect (AHE) and as opposed to AMR, is expected to have a $\sin \theta$ dependence. Deviations from the expected smooth behaviour are typically a fingerprint of strong magnetocrystalline anisotropy. Similar behaviour is observed for both SRO$_{001}$ and SRO$_{110}$ in both $xz$- and $yz$-scans (Fig. 6.11(c), (d), (g) and (h)). In each plot, a maximum transverse resistance is realised whenever the field points along the $+z$ direction ($90^\circ$) and minima in resistance when the field points in the $-z$ direction, as expected. However, the transition between one state to the other is relatively sharp resulting in a deviation from the expected smooth sine dependence. Fig. 6.11(d) and (h) also indicate that for SRO$_{110}$ when the field is aligned in-plane a single sharp transition occurs while for SRO$_{001}$ (Fig. 6.11(c) and (g)) a series of steps are seen spanning a larger range of angles around $0^\circ$ and $180^\circ$.

The transverse resistance ($R_H$), measured perpendicular to the current ($y$) direction, results from the anomalous Hall effect (AHE) and as opposed to AMR, is expected to have a $\sin \theta$ dependence. Deviations from the expected smooth behaviour are typically a fingerprint of strong magnetocrystalline anisotropy. Similar behaviour is observed for both SRO$_{001}$ and SRO$_{110}$ in both $xz$- and $yz$-scans (Fig. 6.11(c), (d), (g) and (h)). In each plot, a maximum transverse resistance is realised whenever the field points along the $+z$ direction ($90^\circ$) and minima in resistance when the field points in the $-z$ direction, as expected. However, the transition between one state to the other is relatively sharp resulting in a deviation from the expected smooth sine dependence. Fig. 6.11(d) and (h) also indicate that for SRO$_{110}$ when the field is aligned in-plane a single sharp transition occurs while for SRO$_{001}$ (Fig. 6.11(c) and (g)) a series of steps are seen spanning a larger range of angles around $0^\circ$ and $180^\circ$.

In addition to the first harmonic signals, we studied second harmonic responses. As well as linear effects, the voltage response also contains higher-order terms due to the nonlinear current-induced response of the system. The spin-orbit torques, for instance, scale linearly with the current and therefore the SOT-related voltage signal is expected to scale quadratically with the current. The effect of current-induced spin-orbit torques on the magnetisation direction can therefore be probed with the second harmonic component of the voltage. It should be noted that also thermal effects scale linearly with current and therefore manifest in the second harmonic voltage signal. Overall, studying the variation of the second harmonic signals as a function of the field orientation allows for identifying current-induced modulation of the magnetisation, whether of thermal or non-thermal origin.

The longitudinal second harmonic data when rotating the field in the $xz$ plane are shown for SRO$_{001}$ and SRO$_{110}$ in Fig. 6.12(a) and (b) respectively. Both graphs reveal strong and clear signals with a $180^\circ$ periodicity. In both cases, the angular dependencies comprise a (series of) vertical jump(s) between the signal maximum and minimum as well as a sloped line between a minimum and maximum. For SRO$_{110}$, a sharp jump spanning only a narrow range of angles occurs whenever the current and field are parallel. For SRO$_{001}$, on the other hand, a series of three jumps

Figure 6.12: Second harmonic angular measurements for SRO001 (left) and SRO110 (right). Data for $xz$ sweeps are shown in (a) and (b) for the longitudinal resistance and in (c) and (d) for the transverse resistance. The $yz$ scans are shown in (e) and (f) for the longitudinal resistance and in (g) and (h) for the transverse resistance. The angle ($\theta$) of the magnetic field ($B$) is indicated with respect to the in-plane direction. $\Delta R$ indicates the change in resistance with respect to when the field is applied out-of-plane. Red and black lines are used to indicate the trace (increasing angles) and retrace (decreasing angles) scans respectively.
and cusps appears between the peak and dip. The transition from peak to dip is still centred around 0°, but spans a wider range of angles of around 60°. On the other hand, no clear signals are observed for either heterostructure when the magnetic field is rotated in the yz plane as seen in Fig. 6.12(e) (SRO_{001}) and (f) (SRO_{110}). This can be due to a too low signal-to-noise ratio or the absence of angular dependence.

The transverse signals for xz plane rotations are significantly different. Both signals appear to have a 360° periodicity, but their shapes differ. For SRO_{001} there is a peak in the signal at 0° and a minimum at 180°. Jumps and cusps are present around a relatively wide range of angles surrounding the situation when the field points in-plane. For SRO_{110}, a rapid change in the signal is seen at 180° and a small jump-like feature appears when the angle is 0°, given that this feature appears consistently between trace and retrace this is not believed to be an artefact.

The transverse signals for rotations in the yz plane yield clear angular dependencies for both samples (Fig. 6.12(g) and (h)). The signal-to-noise ratio is remarkably high for both. Qualitatively, the angular variations are similar to the longitudinal signals in the xz scans. The periodicity of the signals is 180° and the graphs show (a series of) vertical jumps between global dips and peaks. For SRO_{110}, a single sharp jump is observed when the field is in-plane, while for SRO_{001}, a series of jumps spanning a wider range of angles is seen.

### 6.7 Elimination of Thermal Effects

The harmonic Hall measurements include various thermoelectric contributions, Park et al.\cite{54} proposed a method to eliminate major thermoelectric signals. This method works for sufficiently large fields where no hysteresis is seen in the second harmonic signal, as is the case for the data presented here. A current injected into a Hall bar generates a temperature gradient giving rise to thermoelectric contributions. First harmonic signals indicate the equilibrium magnetisation direction and second harmonic signal show SOT-induced magnetisation tilting.

The second harmonic Hall voltage is sensitive to contributions from the Seebeck ($V_S$), Nernst ($V_N$), Ettingshaussen ($V_E$), Righi-Leduc effects ($V_R$), as well as an Ohmic offset ($V_O$). These effects can be removed by taking their symmetries with respect to current and magnetic field into account. This requires four measurements to be taken with different current and field polarities, giving rise to four $V_{IB}^{\pm \pm}$ terms where superscripts $I$ and $B$ refer to the current and field polarities respectively.

\[
\frac{V_{i+}^{++} - V_{i-}^{+-} - V_{i+}^{+-} + V_{i-}^{-+}}{4} = V_{AH} + V_{H} + V_{E}
\]  (6.4)

$V_E$ is usually negligible, allowing for the Hall and anomalous Hall contributions to be isolated.
Figure 6.13: Elimination of second harmonic thermal effects to Hall voltages. Scans taken with a field of 7 T and 1.5 mA on Pt/SRO/STO (001) (left) and Pt/SRO/STO (110) (right). (a) and (b) show the data for the $xz$ scans after eliminating thermal effects using Eq. 6.5. (c) and (d) show the data for the $yz$ scans after eliminating thermal effects using Eq. 6.6.

Similarly for the second harmonic signals: Damping-like SOT geometry ($B \parallel x$):

$$\frac{V_{2\omega}^{++} - V_{2\omega}^{+-} + V_{2\omega}^{-+} - V_{2\omega}^{--}}{4} = V_{DLT} + V_R$$  \hspace{1cm} (6.5)

$V_R$ is a second-order effect and is consequently negligible. Field-like SOT geometry ($B \parallel y$):

$$\frac{V_{2\omega}^{++} + V_{2\omega}^{+-} + V_{2\omega}^{-+} + V_{2\omega}^{--}}{4} = V_{FLT} + V_S + V_O$$  \hspace{1cm} (6.6)

$V_S$ and $V_O$ contribute a constant offset that can readily be subtracted to isolate the effect of field-like torque.

Appendix E, Figs E.10 and E.11 (a) and (e) contain the first harmonic Hall voltages with the four different measurement geometries. Using Eq. 6.4, the thermoelectric contributions were eliminated and the results of this are shown in Figs E.10 and E.5 (b) and (f). Figs E.10 (c) and E.11 (c) show the second harmonic Hall voltages measured using the four geometries for $xz$ rotations. The thermal contributions were removed using Eq. 6.5, giving rise to the graphs in Fig. 6.13 (a) and (b). Figs E.10 (e) and
E.11(e) show the second harmonic Hall voltages measured using the four geometries for $yz$ rotations. Here, Eq. 6.6 was used to remove the thermoelectric contributions, resulting in the graphs shown in Fig. 6.13(c) and (d). This analysis indicated that the line shapes of the first and second harmonic Hall signals are not significantly altered by subtracting the thermoelectric artefacts, suggesting SOT to be the origin.

6.8 Switching Experiments

Figure 6.14: Switching Experiments. (a) Schematic of the experimental setup used. (b) Anomalous Hall resistance measured as a function of field. The red, blue and green lines indicate the up and down magnetised and demagnetised remnant states respectively.

First, an out-of-plane field was applied to measure the Hall voltage as a function of the field and determine the positive and negative remanent magnetisation states; the results of this are shown in Fig. 6.14(b). The red and blue lines indicate the Hall resistance at 0 T when the magnetisation points up and down, respectively while the green line indicates the demagnetised state. An out-of-plane field with a magnitude high enough to saturate the SRO was then applied either up ((c) and (d)) or down ((e)) and removed. A constant AC was then applied (100 $\mu$A, the same value as
used for the Hall loop in Fig. 6.14(a)) and as a function of time the resistance was measured. A Keithley source-measure unit was connected along the long axis of the Hall bar and DC pulses were applied on top of the AC. New pulses were applied after the state was observed to be stable.

Several observations can be made about the data. (i) Current pulses of 15 mA and lower did not affect the Hall resistance \( R_H \), while \( R_H \) decreased ((c) and (d)) or increased ((e)) in a stepwise manner when higher amplitude pulses were applied. (ii) The change in \( R_H \) depended on the amplitude of the pulse, but not on the sign of the pulse. (iii) If pulses of the same amplitude were applied multiple times, only the first pulse affected \( R_H \) and it remained constant afterwards. (iv) If a lower and higher amplitude pulse were subsequently applied, the final state was the same as when only the higher amplitude pulse was applied. (v) The magnetisation always approached the demagnetised state and never switched direction.

These results can have two origins. Firstly, this could be the result of probabilistic switching of a multi-domain ferromagnetic layer. In this case, each pulse affects a portion of the domain in the layer and randomly switches it up or down, resulting in a reduction of the overall magnetisation. Secondly, this could be caused by Joule heating where the current locally heats parts of the layer, causing its temperature to surpass \( T_C \) and its magnetisation to be lost. Joule heating effects could be mitigated by reducing the pulse times.

6.9 Discussion

For SRO grown on STO (001) the XRD results showed that for both the substrate and film (in pseudocubic notation) the out-of-plane axis is [001], and the in-plane axes of the film also align with the equivalent axes of the STO. Similar conclusions can be drawn for SRO grown on STO (110), where again the film axes are aligned with the equivalent substrate axes. As a result, the [110] \(_{pc}\) SRO crystal axes, i.e. the expected magnetic easy axes, for SRO\(_{001}\) and SRO\(_{110}\) lie in different spatial directions. This is confirmed by the magnetic measurements shown in Fig. 6.9(a) and (b) that indicate the easy axis of SRO\(_{110}\) to lie along the film normal, while that of SRO\(_{001}\) is tilted away from the film normal. The combined results of XRD and magnetisation studies are summarised in Fig. 6.9(c) and (d), indicating the unit cell orientations and magnetic easy axes of the different samples. The longitudinal resistance is predominately attributed to AMR arising from conducting through the metallic SRO. It is typically observed that in heavy metal/ferromagnetic metal systems contributions from AMR are significantly larger than those arising from spin Hall magnetoresistance\(^5^5\). AMR is typically composed of two components (crystalline and non-crystalline) that have different microscopic origins. The non-crystalline compo-
The non-crystalline AMR dominates in the \(xz\) scans shown in Fig. 6.11(a) and (c). For SRO\(_{001}\) we observe that the resistance is lower when the current and field are parallel than when they are perpendicular. This is commonly referred to as negative AMR. While it is opposite to what is found in the most common ferromagnets, it has been observed in SRO and several other systems\[58\]. Given that the spin-polarisation of SRO is \(-9.5\%\)[43, 59], the dominance of minority spin transport could be the cause of the negative AMR\[60, 61\].

Deviations from the expected \(\cos^2\theta\) shape are likely the result of anisotropy. In particular, the sharpness of the dips that occur when the field is in-plane indicates this is close to a magnetic hard axis. On the other hand, the broad and flat peaks centred around the out-of-plane directions indicate the vicinity of a magnetic easy axis. The flattened maxima might further indicate that there are multiple easy axes that lie close to, and on either side of the out-of-plane directions with small tilts. Rather than following the field, the magnetisation may jump between these axes.

For SRO\(_{110}\), a more conventional positive AMR is seen, reflecting the fact that the shape of atomic orbitals depends on the direction of magnetisation. In particular, the scattering cross-section is larger when the current and field are parallel giving rise to maximum resistance while the cross-section is smallest when the field and current are orthogonal.

During the rotational measurements conducted in the \(yz\)-plane, in which the angle between the field and current is constant, the AMR is expected to be more sensitive to the crystalline component. For SRO\(_{001}\), flat resistance maxima appear when the magnetisation lies in-plane, and sharp transitions when the field is \(\pm \sim 30^\circ\) away from one of the in-plane directions. The rapid jump in resistance suggests a magnetic easy axis lying perpendicular to the angle, namely oriented approximately 30° away from the film normal. The broad dip spans a similar range of angles as the peaks in Fig. 6.11(a), again corroborating the existence of easy axes tilted away from the film normal.

The sharp dips in SRO\(_{110}\) when the field is in-plane reflect that the in-plane direction is magnetically hard. The easy axis in this case is expected to lie perfectly out-of-plane. The sharpness of the dips in this scan is in contrast to the broad, but flattened peaks, and cannot be explained considering only low-order anisotropy terms. Instead, higher-order crystalline anisotropy terms should be taken into consideration to fully explain the complex AMR.

The transverse resistance measurements for both samples yield similar results
but with some important differences. The expected sine periodicity is observed where the highest resistance is found whenever the field points up (+z) and the lowest resistance when the field points down (−z). This indicates that for both substrates the AHE resistance is positive, consistent with the out-of-plane field scans shown in Appendix Fig. E.2. The shape deviates from that of a perfect sine as a result of the transitions that occur when the field approaches the in-plane direction. The single sharp transition seen at 0° and 180° for SRO\textsubscript{110} suggests in-plane to be a hard magnetic plane and it to be more favourable for the magnetisation to lie out-of-plane. For SRO\textsubscript{001} the transition region spans a greater range of angles and multiple jumps and cusps are seen to occur in this region. This is consistent with a more complex anisotropy and multiple easy axes with a small tilt away from the film normal.

The longitudinal second harmonic data for $xz$ scans (Fig. 6.12(a) and (b)) yields strong signals with similar features for both samples. One of the significant differences is that for SRO\textsubscript{110}, a single sharp jump occurs when the current and field are parallel while for SRO\textsubscript{001} this transition comprises of three jumps and cusps spanning a wider range of angles of about 60° around the in-plane configuration. This is consistent with the first harmonic transverse results in which a series of (SRO\textsubscript{001}) and a single (SRO\textsubscript{110}) jump(s) are seen for the different samples. As mentioned previously, this is likely a signal of the differences in anisotropy, in particular with SRO\textsubscript{110} possessing well-defined uniaxial anisotropy and SRO\textsubscript{001} having multiple easy axes. No longitudinal second harmonic signals were seen when instead the magnetic field was swept in the $yz$ plane, where the field is always perpendicular to the current. The symmetry of the longitudinal $xz$ signal and lack of $yz$ signal is consistent with the presence of anti-damping like SOT, with a small contribution likely due to thermal gradient\cite{53,62}.

The transverse signals for $xz$ plane rotations have a 360° periodicity. The $yz$ plane rotations yield clear angular dependencies with a period of 180°. These signals could originate from SOT, but there can also be a contribution from a vertical thermal gradient. The high signal-to-noise ratio of the second harmonic signals likely originates from spin-orbit torque in both Pt/SRO/STO samples. Section 6.7 utilises the four-direction method proposed by Park et al.\cite{54} to eliminate thermoelectric contributions from the harmonic Hall signals. The line shapes are not significantly altered by this method, indicating SOT to be the dominant origin of the observed signals.

### 6.9.1 Spintronic Memristor

Finally, we discuss potential applications of these SRO/STO systems in the field of neuromorphic computing. When the easy axis is along the film normal, as is found for SRO\textsubscript{110}, an in-plane magnetic field in the direction of the applied current is needed for the magnetisation switching to be deterministic. In SRO\textsubscript{001}, where the
easy axis is tilted, on the other hand, the symmetry is broken with respect to the in-plane SOT; this makes it possible to achieve field-free deterministic switching in this case. When current is applied through the Pt layer, a spin accumulation will be induced through the spin Hall effect. This, in turn, exerts a spin-transfer torque on the magnetic moments of SRO. With a sufficiently high current density this torque can force the magnetisation to align with the spin accumulation direction\cite{15,17}.

Memristors are circuit elements that can show several resistive states controlled by the amount of electric charge. These devices are deemed particularly useful for realising neuromorphic computing, because their behaviour is reminiscent of that of synapses, and they have also been used to emulate neurons. The magnetisation state in these SRO films is continuously tunable and can be used as memristors when included as the free layer in a magnetic tunnel junction (MTJ). Here the state of magnetisation can be read by tunnel magnetoresistance. A proposed device structure for a three-terminal spintronic synapse consists of two transistors and one MTJ device (2T1MTJ)\cite{63,64}. A schematic of the MTJ device is shown on the left hand side of Fig. 6.15. The device consists of an STO substrate/SRO reference layer (RL)/STO tunnel barrier (TB)/SRO free layer (FL)/Pt. Three terminals are required as the read and write paths are electronically separated. The writing path is between terminals $T_1$ and $T_2$, and the read path uses terminals $T_1$ and $T_3$. While isolation of the read and write paths requires two transistors which may be disadvantageous, it enables

Figure 6.15: Schematic of the proposed device structure for neuromorphic spintronic memristors. The device consists of an STO substrate/SRO reference layer (RL)/STO tunnel barrier (TB)/SRO free layer (FL)/Pt. The write path is between terminals $T_1$ and $T_2$ and the read path between $T_1$ and $T_3$. The right side of the figure indicates how the choice of substrate dictates whether the device will show deterministic or probabilistic behaviour.
fast and reliable operation.

A useful benefit of utilising spintronic devices is that the possibility of magnetisation control through both magnetic field as well as current allows both individual devices to be addressed (by current) in addition to all devices simultaneously being set to the same state (using magnetic field). Based on this, we propose a three-terminal synaptic MTJ device in which different non-volatile states can be written by applying an in-plane current. The memory can then be read out by a small out-of-plane current. Introducing the third terminal has several practical advantages. Firstly, it is more energy efficient to separate the read and write paths as writing through the tunnel barrier would require large amounts of current to be passed through an insulating layer, resulting in heat generation. Secondly, by passing large currents through the tunnel barrier it is possible that over time damage occurs, leading to limited endurance.

The right side of Fig. 6.15 indicates how the choice of substrate dictates whether the device will show deterministic or probabilistic behaviour. Using STO (001) substrates we showed that we can realise an easy axis which is slightly tilted away from the film normal, hence breaking the symmetry and allowing field-free deterministic switching in a relatively simple device structure without additional layers. This device could serve as a functional spintronic synapse. In neural networks, synapses typically play the role of representing a weight, which can be viewed as the equivalence of a conductance regulating the flow of information between the two neurons it connects. For a network to function properly, these weights should be non-volatile and able to take on various levels within a range. It should also be possible to controllably change the conductance with an external stimulus. In this case, the weight could be controlled by direct current (DC) pulses - the direction in which the pulse is sent will determine whether it potentiates or depresses the weight of the synapse.

Similarly, when STO (110) is used and the easy axis is oriented fully out-of-plane, the probabilistic nature of switching can be utilised to get different functionalities out of the device. Probabilistic behaviour is becoming increasingly important as stochastic systems are being used for unconventional computing\[65, 66\]. Stochastic MTJs have, for example, been proposed for integer factorisation as an alternative to qubits\[67\] and image segmentation\[68\]. Sourcing a write current will result in probabilistic switching of the free magnetisation, where the probability is determined by the current magnitude\[69, 70\]. This could act, for example, as a non-linear activation function or a spiking spin-neuron in a network. Hence, by controlling the magnetic anisotropy of the system, the same materials can be used to potentially realise both neuronal and synaptic devices.

In order to use such a device utilising SRO as a free layer, an oxide tunnel barrier and a second ferromagnetic PMA layer as the reference layer is needed. Using SRO for the latter as well would be advantageous as again no additional layers are needed.
to induce an out-of-plane easy axis. Such MTJ stacks have been demonstrated by Herranz et al. consisting of uncoupled SRO/STO/SRO stacks on STO substrates in which both SRO layers exhibit PMA[59]. This is also supported by the experimental results presented in Chapter 7. The benefit of this approach lies in the simplicity of the device design compared to conventional PMA devices whose operation relies on a multitude of layers.

6.10 Conclusions

In this chapter, we have demonstrated the ability to control magnetic anisotropy of SrRuO$_3$ ferromagnetic layers by the choice of substrate. The tailored anisotropy can potentially allow both probabilistic and deterministic current-induced magnetisation switching when a Pt layer is added. In light of neuromorphic applications, this gives the possibility to realise two different spintronics memristive devices where the anisotropy controls the device functionality. With perfect perpendicular anisotropy, a device with probabilistic switching could be made which can serve the role of an artificial neuron. By tuning the easy axis to have a slight tilt, the switching can be deterministic and provide synaptic functionality.
Bibliography


