Magnetic anisotropy induced novel phenomena in itinerant ferromagnet SrRuO3 thin films

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Chapter 1

Introduction

1.1 Complex Oxides and Spintronics

The discovery of giant magnetoresistance opened the era of Spintronics, which deals with the interplay between charge transport, magnetic structure, and magnetization dynamics [1, 2]. Spintronics is one of the most attractive and promising fields in condensed matter physics and has opened new frontiers in fundamental research to commercial applications. In this, quantum materials with strong electron correlation have been investigated for emergent phenomena, offering rich potential for technological applications. For instance, the cross-control between ferroelectricity and magnetic order in multiferroic materials (such as $BiFeO_3$) [3], the metal-insulator transition driven by a magnetic field in colossal magnetoresistance materials (such as $LaSrMnO_3$) [4, 5], and the discovery of high-temperature superconducting (such as in cuprates) [6].

The discovery of the quantum Hall effect in 1980 brought semiconductor research to a new era, the famous concept of topology arising from the quantum Hall effect is now a fundamental design principle for quantum materials. To date, many topological quantum materials have been proposed theoretically [7] and several confirmed by experiments [8]. Complex oxide based magnetic materials provide a versatile platform to explore topological spin textures in real space such as skyrmions. As they are protected by topology, skyrmions provide potential prospects in the application of low-power-driven magnetic memory and neuromorphic computing devices, exhibiting a clear advancement over conventional silicon-based integrated chip devices [9]. Nowadays, skyrmion science has been actively developed in different materials systems including bulk compounds, magnetic multilayers, nano-wires/dots [10, 11].

1.2 Resilience and Adaptability: New pathways with Skyrmion bubbles

Magnetic bubbles are cylindrically shaped islands with reversed magnetization that can be set into motion by electric current pulses. They have been discovered in the

1970s and were immediately employed to store digital information in non-volatile memory that can withstand harsh environments. The interest in magnetic bubbles was recently spurred by the discovery that in chiral magnets, their size can be reduced down to a few nanometers, which makes them excellent candidates for novel high-density magnetic memory devices [12]. The magnetic dipoles in the nano-sized bubbles, aka skyrmions, form a knot that cannot be easily unwound. This non-trivial topology gives rise to a skew-scattering of electrons off skyrmions - the so-called Topological Hall Effect. The stability of magnetic bubbles in magnetic thin films with strong spin-orbit coupling extends well beyond the region predicted by the Kooy-Enz model when the film thickness becomes comparable to the cylindrical domain wall width. Small bubbles with skyrmion topology can be stabilized by higher-order magnetic anisotropies [13].

In Spintronics a chief material challenge is related to their implementation as high density, non-volatile memory and logic devices with reasonably good thermal stability and low currents for magnetization switching. The search for materials that exhibit perpendicular magnetic anisotropy and where this can be manipulated by electric field is an active area of research in spintronics, primarily for applications in magnetic memory. Complex oxides and their interfaces provide a rich platform for designing heterostructures with perpendicular magnetic anisotropy and more importantly the ability to tune such anisotropy with electric fields, offering both the flexibility as well as a new design strategy for incorporating such a unit in a practical spintronic device, such as in magnetic tunnel junctions. These can then be driven by current or spin orbit torque for their use as a magnetic memory or as building blocks for neuromorphic functionalities and in the emerging field of Orbitronics.

1.3 $SrRuO_3$ for spintronic and topological phenomena

$SrRuO_3$ is unique as it is the only known 4d transition-metal oxide ferromagnet, where the perfect cooperation between electron-electron correlation and spin-orbit coupling in the $SrRuO_3$ allows for the design and study of different emergent phenomena. To date, plenty of novel physical phenomena have been found in $SrRuO_3$, such as itinerant ferromagnetism [14], magnetic monopoles in momentum space [15], magnon transport [16], signatures of Weyl fermions [17] and Topological Hall effect [18]. Central to this is the octahedral rotation and the control of their magnitude and direction which has been studied both in bulk as well in thin films. The additional flexibility that lattice strain offers in thin films of $SrRuO_3$, in designing different crystalline symmetries has been exploited to the understanding of how octahedral tilts lead to new functional properties, chiefly in the interplay of magnetic states. Different x-ray-based absorption or spectroscopic techniques have been used to study this in $SrRuO_3$ or in conjunction with an antiferromagnet such as $SrMnO_3$ in heterostructures. In particular, the $SrRuO_3$ single layer and heterostructures are perfect model systems to explore topological spin textures, which might trigger a wide range of memory device applications, especially in the design of exchange biased and perpendicular magnetic anisotropy layers which are commonly used in Spin Transfer Torque based MRAMs.

1.4 Motivation and thesis outline

The main aim of this thesis is to explore novel phenomena by manipulating the magnetic anisotropy in magnetic materials. Complex oxides provide an ideal platform in this respect where the competing ground states due to the interplay between charge, spin and orbitals offer such possibilities. For this purpose, we use thin films of $SrRuO_3$ deposited on different single crystalline substrates such as $SrTiO_3$ and $LaAlO_3$ and study the dependence of the magnetic anisotropy of such films with the symmetry and magnitude of the epitaxial strain. We also study the evolving magnetic behavior with thickness, composition, and lattice strain by magneto-transport studies. In this thesis, we comprehensively study the magnetism and magneto-transport (AHE, AMR) in single $SrRuO_3$ and $SrMnO_3/SrRuO_3$ thin films. We believe that our work provides an important contribution to the understanding of such interfaces for future applications by exploiting the complex magnetocrystalline anisotropy intrinsic to these materials and their manipulation by electric and spin-orbital fields.

This thesis is built up in the following chapters, of which a brief overview is given below:

- *Chapter 2* introduces the basic physical concepts needed to understand the work presented in the following chapters. Firstly a general introduction into complex oxides with perovskite is given. Thereafter, basic concepts in magnetism used in this thesis are introduced, followed by the introduction of the magneto-transport effect. Finally, important material properties of $SrRuO_3$ are discussed.

- *Chapter 3* explains the thin film deposition process, characterization methods, and device fabrication which are used for the experiments presented in this thesis. First, the thin film deposition process is discussed, including substrate preparation, pulsed laser deposition, and in situ monitoring by RHEED. Followed by the introduction of characterization methods. This is followed by a description of the steps of the Hall bar device fabrication and the used measurement setups, including an explanation of the lock-in detection technique.

- *Chapter 4* discusses the growth conditions, structures, and magnetic properties of $SrRuO_3$ thin films deposited on different substrates ($SrTiO_3$, $LaAlO_3$). We provide an understanding of the effect of epitaxial strain and film thickness on $SrRuO_3$ thin films. We find that all the $SrRuO_3$ thin films exhibit strong anisotropy and coexistence of high-spin state and low-spin state, and the magnetic behavior of $SrRuO_3$ thin film is sensitive to the epitaxial strain and substrate termination.

- *Chapter 5* investigates the topological spin textures in single $SrRuO_3$ thin films. Using a combination of Hall transport studies and numerical simulations as well as density functional theory, we observe characteristic signatures of the Topological Hall Effect associated with skyrmions. The transport anomalies exhibit an unprecedented robustness to magnetic field tilting and temperature variations. Our numerical simulations suggest that this unconventional behavior results from compact magnetic bubbles with skyrmion topology stabilized by magneto-dipolar interactions and higher-order magnetocrystalline anisotropies in an unexpected region of parameter space.

- *Chapter 6* explores how the interface condition between $SrMnO_3$ and $SrRuO_3$ modifies the magnetic anisotropy, and the evolution of spin textures in the system. We demonstrate that differences in the oxygen vacancies at the heterointerface of $SrMnO_3/SrRuO_3$ can also strongly influence the magnetocrystalline anisotropy in $SrRuO_3$, despite being fully strained by the underlying substrate. Modification of the spin-orbit coupling strength by altering the hybridization of Ru-4d and O-2p orbitals in $SrRuO_3$ leads to a clear evolution of the magnetocrystalline anisotropy from multiaxial to strongly out-of-plane, as manifested in the magneto-transport studies.

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