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# Summary

Spintronics is one of the most attractive and promising fields for future information technologies, in terms of both data storage and logic computing. In Spintronics, the central material challenge is related to their implementation of 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 the magnetism can be manipulated by electric field is an active area of research in spintronics, primarily for applications in magnetic memory. Complex oxides with perovskite structures and their interfaces provide an ideal platform due to the competing ground states arising from the interplay between charge, spin, and orbitals. $SrRuO_3$ (SRO), the unique 4d transition-metal oxide ferromagnet, is an attractive material to design different emergent phenomena due to the strong cooperation between electron-electron correlation and spin-orbit coupling. Furthermore, the tunability of the magnetic anisotropy in SRO-based heterostructures provides opportunity to create unconventional features in the magnetic and magneto-transport properties. This thesis focuses on the study of novel features in SRO thin films and heterostructures via tuning the magnetic anisotropy of SRO.

The ferromagnetism and metallicity in SRO arise from the strong hybridization of the Ru 4d and O 2p bands which enhances the density of states at the Fermi level. Due to the strong correlation between physical properties and crystal structure, the magnetic and electronic properties of SRO thin films are strongly influenced by the $RuO_6$ octahedral distortion. The octahedral distortion can be controlled by strain modification via different substrates and film thicknesses. In **chapter 4**, first, an overview of the effect of epitaxial strain and film thickness on the magnetic properties of SRO is introduced. Followed by a description of the growth details of SRO thin films studied in this thesis. $SrTiO_3$ (STO) and $LaAlO_3$ (LAO), as the most common substrates for complex oxides, are used as substrates in our study to compare

the effect of strain on the magnetism in SRO. Through the study of the structures and the electric properties of SRO thin films with different thicknesses, we notice that structural disorders are unavoidably induced at the early stage of film growth in SRO thin film. Due to the strong electronic correlation in SRO, the micro-structural disorder could play an important role in the magnetism and magnetotransport properties. To investigate the magnetic anisotropy of SRO films, we provide an understanding of the effect of epitaxial strain and film thickness on the magnetic properties of SRO thin films. All the SRO thin films have easy axes close to out-of-plane direction, which is because the epitaxial SRO films are $(110)_O$-oriented and have strong magnetocrystalline anisotropy (MCA) in this direction as introduced in chapter 2. Additionally, we find all the SRO films show coexistence of a high-spin state and a low-spin state, where the high-spin state is formed at the interface induced by the octahedral distortion. Furthermore, we observe a decrease in the saturation magnetization with increasing thickness. As a consequence, the coexistence of high-spin state and low-spin state configurations gives rise to an antiferromagnetic (AFM) coulping of interfacial SRO and softens the magnetic hysteresis, leading to two-step features in the M-H hysteresis. The results in this chapter indicate that the magnetic anisotropy in SRO thin film is canted and the AFM nature coupling of the mixed spin-states could pin the magnetic moment to a certain direction. Furthermore, the study of the magnetic anisotropy energy for SRO, shows the higher-order anisotropy constant still plays an important role, while it is almost negligible in other systems. Based on these features, it provides possibilities to explore noncollinear spin textures in SRO thin films.

When electron flow passes through a noncollinear magnetic texture, a Berry phase is generated and acts as an effective magnetic field to the electron flow, giving rise to an additional contribution to the Hall resistance known as the Topological Hall effect (THE) which can be verified via Hall transport study. In **chapter 5**, epitaxial SRO films on STO (001) substrates with thicknesses around 8.7 nm are selected for Hall effect study. The films are patterned into Hall bars with a channel width of 50 $\mu$m after the structural and magnetic characterization. Systematic Hall effect studies are performed at various temperatures and the titled magnetic field. We observe characteristic signatures of THE in these single SRO films. THE has an important contribution to the Hall resistivity in the materials hosting spin configurations with topological characteristics, such as bubbles and skyrmions. The exploration of THE and skyrmions are recently reported in ultrathin SRO films or SRO/$SrIrO_3$ (SIO) ultrathin bilayers, where the THE and expected skyrmion are mediated by the interfacial Dzyaloshinskii–Moriya interaction (DMI) which is induced by strong spin-orbit coupling (SOC) and broken inversion symmetry at the interface. In this work, however, the relatively large thickness of SRO films and the absence of heavy-metal layers make the interfacial DMI an unlikely source of these topological spin textures.

In addition, the transport anomalies exhibit unprecedented robustness to magnetic field tilting and temperature variations. We invoke the hitherto ignored higher-order MCA terms in addition to the long-ranged magnetodipolar interactions and ascribe them to magnetic bubbles with skyrmion topology. Our numerical simulation studies show that the skyrmion crystals induced by magnetostatic interactions have a much greater stability region than it was previously thought. Our method allows us to optimize the shape of skyrmions forming the crystal, in contrast to the (Kooy-Enz) model of magnetic bubbles with sharp boundaries. At the same time, our method makes it possible to circumvent the time-costly micromagnetic simulations, which allowed us to construct detailed phase diagrams in these material systems. These new insights not only advances the field of skyrmions based physics but are timely and provide significant opportunities to tailor such interfaces for future applications by exploiting the complex MCA intrinsic to these materials and their manipulation by electric and spin-orbital fields.

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 by different strategies. The strong temperature and angular dependence of the multiaxial anisotropy indicate the complex nature of MCA in SRO. On the other hand, it provides opportunities to design desirable anisotropy with proper engineering. The magnetic ordering in $SrMnO_3$ (SMO) can be dramatically changed from AFM to ferromagnetic by adjusting the growth conditions, providing a tunable exchange coupling at the interface between SMO/SRO, while SRO has strong MCA which tends to stay normal to film surface. But in practice, the unexpected disorder or inhomogeneity gives rise to multiaxial anisotropies. Such SMO/SRO interface attracts our interest. In **chapter 6**, we investigate how differences in the oxygen vacancies at the heterointerface of SMO/SRO strongly influence the MCA in SRO. We design two different SMO/SRO interfaces by tailoring the growth conditions, such as adjusting the oxygen pressure in SMO. A systematic study of the structure, magnetization, and angle and field-dependent magnetotransport is performed. We find that a higher oxygen pressure during the SMO growth leads to a strong uniaxial out-of-plane anisotropy of SRO, while the oxygen deficient in SMO favors a tilted anisotropy in SRO. By comparing the bilayer results with the single SRO film grown directly on the STO substrate, we conclude that the modification of the SOC strength by altering the hybridization of Ru-4d and O-2p orbitals in SRO leads to a clear evolution of MCA from multiaxial to a strong out-of-plane, as oxygen vacancies in SMO are reduced. As expected, the THE signal is observed in the tilted anisotropy bilayer sample, the existence of the THE signal shows great agreement with the results in chapter 5. The strong uniaxial perpendicular anisotropy in SRO, however, no THE has been found. Nevertheless, such a bilayer system is a promising building block for practical spintronics devices where the MCA can be tuned and magnetization can be controlled, providing a new design strategy for memory and computing applications.

Overall, in this thesis, we comprehensively study the magnetism and magneto-transport in single SRO and SMO/SRO thin films. We observe noncollinear spin textures in the multiaxial anisotropy SRO films. On the other hand, we obtain strong uniaxial perpendicular anisotropy in SRO by adjusting the interfacial conditions. We believe that our work provides an important contribution to the understanding of such interfaces for future applications by exploiting the complex MCA, intrinsic to these materials and their manipulation by electric and spin-orbital fields.