Spin transport across oxide semiconductors and antiferromagnetic oxide interfaces

Das, Arijit

DOI:
10.33612/diss.150692255

Publication date:
2021

Summary

Strong correlated interplay of spin, orbital and lattice degrees of freedom in complex oxide materials, both in their bulk as well as at their heterointerfaces lead to emergent phenomena, that are often unlike those found in other material systems. Such an interplay is brought by a cumulative interaction of the lattice distortion, induced strain and different type of symmetry breaking, enabling effective control and engineering of heterointerfaces. This has led to the observation of unconventional electronic transport, magnetism, ferroelectricity and superconductivity at such heterointerfaces.

This thesis studies functionally important complex oxide platforms for spin transport and explores, (i) Integration of non-volatile memory functionalities on complex oxide semiconducting Nb-doped $SrTiO_3$ (Nb:STO) for complementary memory and logic operation, (ii) Antiferromagnetic insulating films of $SrMnO_3$ (SMO) are studied for the first time, with an outlook to integrate them in antiferromagnetic spintronics based on complex oxides.

An important requirement for advancing conventional semiconductor spintronics is the possibility to control spin manipulation across the semiconducting channel by electric fields. However, this has remained elusive in conventional semiconductors such as in Si, due to the weak spin orbit coupling intrinsic in them, preventing effective manipulation of spin transport by electric fields using an applied bias . The broken inversion symmetry at the surface of Nb-doped $SrTiO_3$ (Nb:STO) and the strong non-linear variation of the dielectric permittivity lends itself an electronically rich platform where such a manipulation can be studied by utilizing carefully tailored

spin injection interfaces. The analysis of electric field manipulation of the spin transport parameters in Nb:STO constitutes a major part of this thesis.
In the second theme of the thesis, antiferromagnets are studied on undoped $SrTiO_3$ as substrates. Antiferromagnets are rapidly evolving as a league of their own in spintronics. However, material choice and device design are key aspects that needs attention to further their development for use in mainstream technologies. Materials that provide additional flexibility, such as in the tunability of magnetic ordering, provides an important advantage. In this regard, the rich materials landscape, inherent to correlated oxides has not been probed, in spite of the fact that several important emergent phenomena have been discovered in these materials or across their heterointerfaces. Another challenge in antiferromagnet spintronics lies in the adaptability of the commonly employed techniques used to study spin transport using ferromagnets. In this thesis, a suite of spin transport based techniques , complemented by bulk magnetization studies are presented with some important conclusions and outlook that sets the stage for their study both by spin currents as well as by electric fields.

In Chapters 1 and 2 the material systems, namely, $SrTiO_3$ and $SrMnO_3$, are introduced along with the required theoretical concepts related to spin injection and detection as well as phenomena based on Spin Hall Effect (SHE) such as Spin Hall Magnetoresistance (SMR) and Spin Seebeck Effect (SSE).

In Chapter 3, the charge and spin transport across Schottky interfaces with Nb:STO are investigated. The Schottky interface of Nb:STO is replete with oxygen vacancies, allowing a metastable state in the conduction band of STO, hence leading to a memristive behavior across Ni / Nb:STO. Moreover, the non-linear variation of the dielectric permittivity of STO with electric field and temperature controls the bending of the conduction band. Information about the band bending and the associated built-in electric field is modelled using electrostatics that shows an electric field variation of 300 kV/cm – 1200 kV/cm. The electronic transport

across Ni /Nb:STO is studied with different doping concentration of Nb^{5+}. With increasing doping concentration, the transport mechanism changes from thermionic emission to thermally assisted tunneling. Increasing tunneling transport is favorable for spin dependent tunneling across the semiconducting interface. Tunneling Anisotropic Magnetoresistance (TAMR) signals are observed across the Schottky interface. Their bias dependence is brought about by the modulation of the built-in electric field , that in turn, modulates the Rashba Spin-orbit field. From these observations, it is concluded that: (i) the tunneling densities of states largely depend on the band structure alignment of 3d-orbitals between Ni and Nb:STO, (ii) the bias dependence relies on the balance between the depletion width and doping concentration of the semiconductor, that provides a controlled bending of the conduction band in STO.
An earlier work in our group on Co/Nb:STO interfaces showed a TAMR vaue of 1.6% and was the largest TAMR reported at room temperature. This value is also an order larger than the highest TAMR reported in this thesis. This makes Co/STO interface interesting to explore further. Earlier researchers reported on an inversion of Tunneling Magnetoresistance (TMR) across Co-STO interface in a magnetic tunnel junction (MTJ), and attributed it to the cleanliness of the interface, but the role and impact of a self-oxidized CoO interface on the magnetoresistance studies were not explored. In the latter part of chapter 3, the observation of magnetoresistive signals with Co/CoO electrode on Nb:STO is discussed. The exchange pinning of Co and CoO magnetic moments at low temperatures indicates a competition between incomplete rotation of Co/CoO moments due to the exchange spring effect and field dependent rotation of Co moments. At room temperature, i.e. above the *Néel* temperature of CoO ($\sim 290K$), quadratic responses of TAMR signals of Co/Nb:STO are observed. This work also highlights that a 3T geometry can be used to study magnetoresistance effects arising at such exchange biased interfaces.

In chapter 4, a careful design of the spin injection interfaces using a Ni ferromagnet and a thin tunnel barrier (0.7 nm) of AlO_x is shown to lead to a sizeable built in electric field at the Schottky interface which is

very effective in tuning spin lifetime in Nb:STO. Usually the presence of a Schottky barrier is detrimental to spin transport, but it is shown that when cleverly engineered, this can lead to a new understanding of spin transport in semiconductors. The experiments are performed using 3T geometry techniques. Although demonstrations of spin injection and detection in Si and other semiconductors were done using the 3T geometry, this geometry has also attracted some undue controversies making researchers wary of publications using this geometry. In this chapter, it is demonstrated that the 3T geometry can be very well exploited to explore new features in spin transport by increasing the material and device parameter space and by careful engineering the spin injecting interface with Nb:STO. By careful and systematic analysis, it is found that the line shape of the spin voltage is a superposition of two competing magnetoresistance effects that evolve in opposite ways with temperature and applied bias. The spin voltage lineshape comprises of spin injection signal superposed with tunneling anisotropic magnetoresistance (TAMR). Such a demonstration is possible due to the concerted effects of (i) the non-linear response of the dielectric permittivity with temperature and applied bias that is intrinsic to Nb:STO (ii) the modulation of the Rashba spin-orbit field at such interfaces with broken inversion symmetry and iii) the effects of i) and ii) on engineering the potential landscape for spin transport.
This work presents new opportunities in electric field control of spin transport in semiconductors by exploiting the potential of 3T geometry and expanding the parameter space to explore new directions in semiconductor spintronics.

In chapter 5, The magnetic properties of insulating manganite thin films of $SrMnO_3$ are introduced for plausible spin and magneto-transport application. The two commonly used electrical (spin) transport schemes for investigation of the magnetization and magnetic ordering in a magnetic insulator (MI) are Spin Hall Magnetoresistance (SMR) and Longitudinal Spin Seebeck Effect (LSSE) studies. Recently, many researchers have adopted a novel way to simultaneously detect the SMR and SSE responses by utilizing the current applied across a heavy metal (HM) bar that generates, (i) a spin current due to Spin Hall effect (SHE) (ii) Joule heating. In a HM/MI hybrid system, both the spin current and the temperature gradient due to Joule heating, propagate across the HM/MI interface creating, (i) a relative orientation of the spin accumulation in the HM and the magnetization at the surface of the MI, (ii) magnon excitation in the bulk of the MI due to the temperature gradient. These effects, in turn, produce a change in the electrical voltage in the HM layer due to the inverse-SHE (ISHE). The voltages are obtained as 1st (SMR) and 2nd harmonic responses with a lock-in amplifier, where the SMR responses are linearly dependent on the current bias and SSE responses varies quadratically with the current bias indicating a thermal effect. Such complementary spin transport techniques have been employed earlier to probe the magnetic ordering in ferri/ferromagnetic, antiferromagnetic and spiral magnetic systems. However, in systems like $SrMnO_3$ (SMO) thin films, the introduction of ferroelectric ordering induced by strain can lead to transformation of the antiferromagnetic ground state to a ferromagnetic one. Such a strain can be achieved by deposition of SMO films on substrates that induce tensile strain like $SrTiO_3$. Moreover, coexisting magnetic phases within a film with varying bulk and surface magnetic ordering can be brought about varying the oxygen stoichiometry by adopting different growth strategies.

In this work, the magnetic characteristics of thin films of insulating SMO are studied using three different complementary techniques, (i) Magnetization measurements from SQUID, (ii) SMR and (iii) SSE. In this work, the role of the correlated interplay of the strain quotient and oxygen vacancy that is tuned in the two samples by adopting different thickness and growth conditions are highlighted. The simultaneous detection of the SMR and SSE in the two samples reveals a predominant ferromagnetic interaction at all measured temperatures, this is further supported by the presence of magnetic hysteresis. On the other hand, from the SSE responses that fingerprint the bulk magnetic ordering in a MI, a gradual canting of the antiferromagnetic sublattices with increasing field strength is thought to be responsible for a competing magnetic exchange laterally across the SMO films. A temperature dependent study of the SSE responses in the thin films reveals an enhancement in the signals at lower temperatures, possibly

hinting at a strong magnon-phonon interaction that is mediated across the surface and bulk of the SMO films, when they are strongly coupled by a competing magnetic exchange also evinced by the temperature dependent magnetization measurements using SQUID. Significant efforts have been made to rule out other possible effects by using different configurations. This study is the first of its kind to highlight the presence of such competing magnetic ordering in thin films of SMO and manganites in general using the novel spin transport techniques of SMR and SSE.
This work also inspires the scope of tuning different parameter space in complex oxide materials for viable spin transport measurements and also broadens the scope of SMO as the material for future antiferromagnetic spintronics.
$SrMnO_3$ can be tuned to exhibit different magnetic ordering, prominent among them being a G-type AFM, where both the intra and the inter-sublattices are cancelled by each other. This has been used in a heterostructure comprising of $SrMnO_3$ with $SrRuO_3$, the latter being an itinerant ferromagnet in the complex oxide family. The thesis looks at the efficacy of exchange bias via Dzyalonshinkii-Moriya interaction (DMI), in such FM/(nominal) AFM system by studying the magnetotransport properties. In **chapter 6**, integrating thin films of SMO with SRO, is found to show hump like features typically observed in Topological Hall effect (THE) studies, due to the electron scattering from topologically protected skyrmion-bubble textures across the interfaces. By tuning the exchange interaction, utilising two different thickness of SMO films, we observe a magneto-transport behaviour that shows visible differences in their THE response, indicating the role of the DMI and zero-magnetostatic stray field induced magnetic textures.
The flexibility in the magnetic phases in the SMO films and its polar properties can be utilized in such exchange bias systems for further study of electric field driven effects of magnetization textures in such heterostructures.