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Chapter 1

Introduction

1.1 Spintronics

Downscaling the transistor size has been an extremely effective method to increase computational power. In the early 1970's a CPU had around 2000 transistors while modern day CPU's have several billion. As the industry approaches transistor sizes where quantum mechanical effects such as charge tunneling start influencing their behavior, the classical design of such transistors starts encountering enormous problems. These include cross-talk between neighboring transistors and increase of leakage current, increasingly larger power consumption, thermal dissipation and difficulty in the realization of viable lithography technology to realize the smaller feature sizes. To circumvent these problems many workarounds are explored such as using FinFET's or higher mobility semiconducting channels based on Ge or III-V semiconductors. These workarounds do not solve the afore mentioned problems but simply shift them to a later time period. This is also evident when realizing that the energy needed to power all these processors increases extremely fast simply because more and more processors are being used in daily life. It seems clear that in the long run a transistor based on different physics is needed, not only to realize the continuation of increasing computational power but also to significantly reduce energy consumption.

Many different technologies are proposed to replace or complement conventional CMOS [6] and efforts are made to predict their viability to indicate which technologies are most promising [7, 8]. One of these is spintronics, which encompasses a broad class of technologies where the spin state of a charge carrier is used to perform logic operations or store information. Spintronics has been extremely successful in the area of data storage where the discovery of giant magneto-resistance led to a large increase in the data storage density of hard disks. Recently, the realization of Spin Transfer Torque Magnetic Random-Access Memory (STT-RAM) opens the way for low power consumption and high density non-volatile random access memory.

Driven by these successes there has been significant interest to control the spin state inside semiconductors to realize spin transistors as first proposed by Datta and Das [1]. This would allow the realization of spin based logic which is expected to lead to more efficient and faster operating devices with lower power consumption. The four main reason for spintronics can be summarized by:

Non-volatile nature: The majority of spintronic device concepts preserve their state when the power is switched off (non-volatile), opening paths to reduce (static) power consumption.
Switching speed: Switching the state variable (spin, magnetization) can be realized with time scales towards fs (THz), much faster than CMOS.[1]

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[1] Note that since spin and charge are linked the electron velocity is also important as well as the fact that there is charge flow through interconnects. The related RC times of those components might form a bottleneck for significant performance benefits on IC level.

Switching energy: Charging of a CMOS gate capacitor requires a high supply voltage (V_{dd}) due to the switching of a potential barrier many times higher than the thermal energy k_bT. Such a large potential barrier is not needed in spin logic thus allowing a much lower supply voltage. Since the intrinsic energy of the device is $E_{int} = C_{dev}V_{dd}^2$ much lower energy consumption is benchmarked [2].
Monolithic 3D integration of logic and memory: Vertical stacking of conventional CMOS with spin based logic and memory (which require only low processing temperatures) allows a huge increase in IC packing density as well as alleviating the logic-memory communication bottleneck[2][3–5].

The successful implementation of such a device depends on four aspects: the injection of a sizable spin accumulation into the semiconducting channel, spin propagation along the channel, manipulation of the state during propagation and finally detection. Therefore the first step towards the realization of semiconductor based spintronics is to electrically inject (and detect) a spin accumulation inside the semiconductor. Due to fundamental issues, such as the conductivity mismatch between conventional ferromagnets and the semiconductor channel [9], the formation of large Schottky barriers and complicated interfacial structure and chemistry [10, 11] this is by no means an easy task.

The first reports of spin injection into a semiconductor made use of electrical injection and optical detection [12–14]. A lot of progress has been made in the last 10 years and several groups have reported all-electrical spin injection, transport and subsequent detection in Si, Ge and GaAs using four-terminal non-local geometries [15–17]. Such a non-local geometry is ideal for investigating spin transport in the non-magnetic channel since it allows measuring spin transport without a charge current background present. Furthermore, electrical spin transport and coherent manipulation of hot-electrons in undoped silicon was also demonstrated [18]. However, the fabrication of the aforementioned device geometries is complicated.

Recently an alternative geometry was proposed which removes the necessity of spin transport and only focuses on the spin injection and detection underneath a single contact [19]. This geometry is generally referred to as the three-terminal geometry and can effectively be seen as a limit of the non-local four-terminal geometry where the inner two contacts have joined and form a single contact. In such a geometry the measured signal also contains a part related to the charge current i.e. the charge and spin currents are no longer separated as in a non-local four-terminal measurement. Such a biased spin injection/detection contact is very likely to be an important part of any commercially viable spin transistor. The three-terminal geometry thus allows studying spin injection and detection at a biased interface. It also has other benefits such as, simpler fabrication and investigating the effect of changing the contact size over several orders of magnitude. As shown in Ref. [20], as well as in this thesis,

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[2] CPU stalling due to unavailability of data

it also allows investigating the spin accumulation in materials with very low spin lifetimes and spin diffusion constants, not accessible by non-local geometries. Many different research groups have utilized this geometry to show spin injection and detection in a myriad of semiconductors [21]. A more in-depth discussion related to the use of three-terminal spin injection contacts can be found in Ref. [21, 22].

1.2 Complex oxides and (doped)-$SrTiO_3$

An important consideration for the realization of beyond-CMOS electronics is the material system from which to realize this. One of the promising classes of materials, not only for spintronics but more broadly for electronics as a whole, is complex oxides [23]. Complex oxides have very rich physics leading to materials exhibiting a wealth of properties such as piezoelectricity, ferroelectricity, multiferroicity, ferromagnetism, high-T_c super conductivity, large electroresistance and the formation of high mobility 2-dimensional electron liquids (2-DELs) at the interface of oxide insulators and $SrTiO_3$ [24–29]. As such, complex oxides are promising candidates for many novel applications such as resistive RAM, ferroelectric RAM and memristors [30, 31]. Complex oxides have already shown desirable properties for the realization of spintronics such as near 100 % spin polarization [32] and colossal magneto-resistance [33, 34].

An important step will be to investigate the suitability of complex oxides as spin transistors. The properties of complex oxide 2-DEL systems are compelling for spintronics because they exhibit a wealth of properties which are strongly tunable [35]. For instance, 2-DELs are of interest for a Datta-Das type spin transistor since there is evidence for strongly electric field tunable Rashba Spin-Orbit Coupling (SOC) [36, 37]. This is a real boon because of the following issue: materials with long spin lifetimes tend to have low SOC while materials with large SOC exhibit short spin lifetimes, however SOC is needed to manipulate the spin state. When the SOC strength can be strongly tuned, using electric fields, it might be possible to only switch it on when needed. However, much of the physics of complex oxide 2-DELs is not yet well understood and exploration of spin injection is complicated. This is especially so because there are many reports of possible magnetism in the system which would be detrimental to spin transport [38–41]. Instead, an oxide semiconductor such as doped $SrTiO_3$ can be a very suitable candidate for spintronics and forms a good testbed to explore the possibility of spin injection in complex oxides.

Semiconducting n-doped $SrTiO_3$ is in many ways different from conventional semiconductors such as Si, Ge and GaAs. Electronically n-$SrTiO_3$ is very rich in physics, which originates from several intrinsic aspects of the material. Firstly, the n-$SrTiO_3$ conduction bands are derived from the Ti d-orbitals which are much more localized compared to the p-orbital derived conduction bands of conventional semiconductors. Secondly, it is a quantum paraelectric [42]: when reducing the lattice temperature SrTiO$_3$ is driven towards a ferroelectric phase transition but never becomes ferroelectric due to quantum fluctuations. As in normal ferroelectrics, where the relative permittivity ϵ_r diverges close to the phase transition, a large increase in ϵ_r of SrTiO$_3$ is observed when reducing the lattice temperature. However, ϵ_r of SrTiO$_3$ saturates at around 4 K where quantum tunneling sets in, suppressing the formation of a ferroelectric ground state.

The large and strongly temperature dependent permittivity has important consequences for its electronic properties among which are: the absence of carrier freeze-out of hydrogenic donors and a strongly temperature dependent electron mobility [43, 44]. Unfortunately, commercial SrTiO$_3$ single crystals are known to contain high concentrations of unintentional acceptors, among which Fe [45, 46]. This not only prevents the growth of n-SrTiO$_3$ films with a low carrier density and a high electron mobility but the magnetic impurities can significantly reduce the spin lifetime. However, Son et al. showed that high quality thin films of n-SrTiO$_3$ can be grown by molecular beam epitaxy with proper growth conditions. Temperature dependent studies of the electron mobilities of these films have provided much insight into the scattering mechanisms of the conduction electrons [47]. On top of this n-SrTiO$_3$ has a complex electronic band structure which becomes even more complicated at an interface or surface [48].

From the structural viewpoint SrTiO$_3$ also has a lot to offer since it allows the epitaxial growth of many complex oxides on it. This eases the integration of many complex oxides with complementary properties, for instance providing integrated memory functionality along with the logic functionality of the SrTiO$_3$ channel itself. Significant progress is being made to realize integration of SrTiO$_3$ with Si, allowing the advantages of integration with the mature silicon technology while introducing the novelty of complex oxides [49–52].

As a first exploration of n-SrTiO$_3$'s suitability for spintronic application this thesis explores spin injection and detection in commercially available Nb-doped SrTiO$_3$ crystals. Clearly these are not the most favorable in terms of electronic properties or purity but provide an easily accessible first glance into the electron spin physics in this material. Due to the complicated nature of spin injection into semiconductors as well as the absence of a decent estimation of the spin relaxation length in n-SrTiO$_3$ a three terminal geometry[19, 21] is employed throughout this thesis to study the properties of the spin accumulation underneath the injection/detection contacts. The three terminal geometry, although providing many advantages over other methods, has two significant complications: 1) due to the presence of a local charge current along with the spin accumulation/current the analysis of the obtained junction response can be significantly more complex and 2) the presence of a charge current can lead to magnetoresistance effects other than that related to spin accumulation. The first complication can also be seen as a boon as it can reveal interesting physics related to spin accumulation, albeit more difficult to analyze and/or explain. The second complication is a significant disadvantage especially when it is not possible to isolate or distinguish the spin signal from spurious magnetoresistance effects. The next section is meant as an introduction to the most important standing issues in three terminal spin injection measurements.

1.3 Standing issues in three terminal spin injection

Recently it has become clear that the local three terminal geometry can be very prone to other magneto-resistance (MR) effects which can be hard to discern from the Hanle effect (spin dephasing)[53–58]. Unfortunately this leads to a very complicated scenario where significant care has to be taken in order to at least establish that the observed behavior is inconsistent with known spurious effects. Currently there is a heavy debate, both on experimental and theoretical grounds, to understand what exactly is measured using such a three-terminal geometry. Questions are raised how to interpret the data and if this relates to interface effects or reflects, at least partially, the spin physics in the semiconductor bulk. Note that not all three terminal spin injection devices yield results which are inconsistent with either 1) theory or 2) measurements using other techniques. There have also been investigations where spin injection was shown in the same device, using three terminal measurements in combination with another technique such as: four terminal detection [59–64], optical detection [65, 66] or injection via ferromagnetic resonance [20, 67, 68]. Also, devices which showed behavior intimately linked to spin accumulation such as: the presence of dynamic nuclear polarization [69–72], strongly temperature or doping density dependent spin lifetimes [19] have been reported. To provide the reader with a balanced view over the current understanding of three-terminal spin injection measurements I will shortly discuss the most important issues related to three terminal spin injection. This also provides a solid ground for interpreting the work described in this thesis.

The three most important observations which raised doubt about the origin of the spin signals measured in three terminal devices are:

The amplitudes of the spin signal After the initial success of three-terminal spin injection [19], subsequent experiments by many different groups showed that the MR signal size can be many times larger than that expected for spin accumulation in the linear regime [21, 73, 74]. To explain this, a model which assumed spin accumulation in localized states near the interface was proposed [73]. However, Jansen et al. pointed out that such an enhancement can only occur if several conditions are fulfilled and hence can not easily explain the widely observed enhanced signal size [75]. Additionally, experiments were performed in devices well outside the necessary conditions range and still showed a significant enhancement [65]. It was also shown that the MR size has an unexpected scaling with the junction interface resistance; the larger the interface resistance the larger the spin signal amplitude. This was observed in devices where the tunnel barrier thickness was systematically increased [76, 77], or by showing strong correlation of the non-linearity of the junction resistance and the spin signal amplitude with bias [78]. Although it was argued that the strongly non-linear transport could result in such correlations [78], it was shown that it is not a likely explanation for the wide spread observation of this behavior [79]. On top of this it was observed that these devices also show a strong increase of the spin signal amplitude with reducing temperature. Based on these observations it seems clear that either a crucial part of the theoretical understanding of the amplitude of the spin accumulation in such a three terminal geometry is missing or the observed spin signal does not originate from spin accumulation.

The 'inverted' Hanle effect In many experiments it was observed that applying an in-plane magnetic field resulted in a Lorentzian shaped increase of the junction voltage which is referred to as the 'inverted' Hanle effect [80]. This is unexpected since the in-plane field, in these experiments, is along the spin quantization axis and should not affect the spin accumulation. To explain this, a theoretical framework involving local magnetostatic stray fields, which partially dephase the injected spins and hence reduce the spin accumulation, was invoked [80]. The applied magnetic field acts on these local stray fields by aligning them, resulting in an increase of the spin accumulation. Although this model accurately describes the observed behavior it is not straight forward to provide direct conclusive evidence that this mechanism is indeed responsible for the behavior.

The low spin lifetimes One of the most important parameters which can be obtained from three terminal Hanle measurements is the spin lifetime of the injected charge carrier in the non-magnetic material. Many experiments reported spin lifetimes which were much lower than expected [21, 22]. As such it is extremely important to understand what the three terminal spin lifetime represents: is it the spin lifetime intrinsic to the semiconductor bulk, the spin lifetime in the semiconductor limited by the spin injection/detection contact or the spin lifetime of certain interface states? The presence of the 'inverted' Hanle effect would indicate that the spin lifetime is limited by magnetostatic fields (due to contacts, defects, impurities etc.) and provides an explanation for the low spin lifetimes. In this context it would make sense that the intrinsic spin lifetime is measured when no 'inverted' Hanle effect is observed. Such a condition is generally fulfilled in FM/SC Schottky contacts where spin lifetimes above 1 ns are frequently observed. However, in many of these devices a spin signal amplitude which is much larger (or sometimes much smaller) than theoretically expected has been measured (see chapter 5.2).

An alternative explanation of three terminal Lorentzian MR signals: impurity assisted tunneling magneto resistance

Due to these observations, efforts where made to investigate the observed anomalous behavior of the spin accumulation. To investigate this Txoperena et al. employed a three terminal geometry to determine the spin lifetime in two metals (Al and Au) which have very different spin lifetimes [53]. This lead to an intriguing observation, the measurements showed that the spin lifetime is the same in both Al and Au. The amplitude of the spin signal also showed anomalous scaling with temperature and interface resistance. They concluded that these observations are inconsistent with spin accumulation in the metals and hence some other unknown effect must be at play. Recently they provided a theoretical explanation, with corroborating experimental support, for the observed response in the metal junctions [54]. It was shown that transport via defects in the tunnel barrier, under certain conditions, could lead to a magnetic field response which strongly mimics the field response expected for a Hanle measurement. Interestingly, they showed that no FM is needed for the observation of this effect which they dubbed as impurity-assisted Tunneling Magneto Resistance (iaTMR). It also provides a possible explanation for all three anomalous observation discussed in the previous paragraphs: the large spin signal amplitude, the low and similar spin lifetimes and the 'inverted' Hanle effect. Observation of iaTMR is also reported for $LaAlO_3/SrTiO_3$ [55] and Nb-$SrTiO_3$ [57] based structures. A recent review article discussing three terminal spin injection and the iaTMR effect can be found in Ref. [58].

However, the model has an extremely important caveat: the iaTMR effect is only present when defects are present in the tunnel barrier. As was shown by Txoperena et al., no magnetic field effects were observed in well oxidized barriers. Only when defects were intentionally introduced in the oxide tunnel barrier a significant Hanle like magnetoresistance effect was observed (the iaTMR effect). In principle, Jansen and co workers had also checked for such anomalies by performing three terminal spin injection experiments where a thin non-magnetic metal (NMM) layer was inserted in between the FM and tunnel barrier [74, 77]. They did not find any Hanle like response. Recently, other groups have probed for the iaTMR effect in well oxidized Cu/SiO_2/n-Si junctions [81] or CoFe/Ta/SiO_2/n-Si junctions [82] and did not find any signature of Hanle like effects in a very large set of samples. They did however observe Hanle and 'inverted' Hanle responses when replacing Cu with CoFe or when removing the Ta layer.

Additionally, the iaTMR effect vanishes at a bias voltage such that $eV < kT$ holds, as mentioned and observed in Ref. [54]. However, there are many three-terminal spin injection devices where Hanle effects are still observed while in this regime ($eV < kT$) such as in Ref. [83, 84] as well as in the work in this thesis. As pointed out by Jansen et al. in Ref. [79], spin injection devices which use thermally driven spin injection via Seebeck spin tunneling are also not compatible with the iaTMR model [85–87]. This strongly suggests that the iaTMR mechanism does not play a dominant role in many of the three terminal spin injection experiments which do show anomalous behavior. In the light of these considerations it is of great interest to see if reports of three terminal spin injection devices exist which can not be explained by both iaTMR and spin accumulation theory. It turns out that these are indeed observed and are discussed in detail in chapter 5.2.

The above discussion shows that the great simplicity with which spin injection can be explored using the three terminal geometry is significantly hampered by the lack of current understanding of the measured response and to discern it from spurious effects. In my opinion there is significant evidence that the iaTMR mechanism can not explain all the observations. Therefore, it seems very plausible that currently unknown physical mechanisms are at play at the spin injection interface. Unraveling this will be important not only for understanding three terminal spin injection but for spintronics as a whole because any all-electrical spin transistor will incorporate such biased spin injection contacts.

1.4 This thesis

As mentioned in the previous section the three terminal geometry provides both advantages and disadvantages. The issues as described in the previous section were raised during the period in which the research for this thesis was performed. As such the results in this thesis are discussed in the light of iaTMR and observed to be complicated to explain within this framework. The results in this thesis are interpreted within the framework of spin accumulation. The aim of this thesis is to explore the possibility to realize spin injection in a complex oxide semiconductor. The choice for the complex oxide's is due to their rich physics which provides many avenues for the realization of novel devices for beyond-CMOS technology.

A brief summary of each of the following chapters is presented below.

Chapter 2 introduces the basic theoretical concepts which are needed to understand the experimental and theoretical work described in the following chapters. It deals both with the charge transport at interfaces with a Schottky barrier as well as the basic principles of spin injection and detection in semiconductors. It also discusses spin-orbit coupling, which plays an important role in this thesis, and some of its important consequences such as spin relaxation and tunneling anisotropic magnetoresistance (TAMR).

Chapter 3 presents experimental studies of spin injection contacts comprising of

Co/AlO_x on Nb-$SrTiO_3$. Here we try to understand the significant changes observed in charge transport when the AlO_x thickness is changed. An electrostatic model is introduced to simulate the potential landscape at such contacts and is used to explain the observed experimental behavior.

Chapter 4 shows the study of spin injection in Nb-$SrTiO_3$ at room temperature. It reports the first observation of electric field tunable spin lifetimes in a three terminal geometry by controlling the Rashba Spin-Orbit Coupling (SOC) strength by altering the built-in potential. Additionally, it is shown that the electroresitive effect in the junction allows non-volatile control over the SOC strength.

Chapter 5 discusses the observed behavior of the devices studied in chapter 4 in more detail. It discusses the influence of using different fit functions on the extracted spin lifetime and spin voltage anisotropy, the implications of TAMR, the observation and behavior of the 'inverted' Hanle effect, the amplitude of the spin signal and an overview of three terminal spin signal amplitudes reported in literature using Schottky spin injector/detector contacts.

Chapter 6 shows the simultaneous presence of both a large TAMR effect and electroresistance in Co/Nb-$SrTiO_3$ Schottky junctions. The bias dependence and the large TAMR amplitude are discussed in the light of the large relative permittivty ϵ_r of Nb-$SrTiO_3$ and the $3d$-orbital character of the conduction band. It is shown that the insertion of a 7 Å thick AlO_x layer strongly suppresses the TAMR amplitude.

Chapter 7 presents temperature dependent studies of the spin injection devices discussed in chapters 4 and 5. It reveals a systematic inversion of the sign of the spin signal when reducing the temperature below $\sim 150\,\mathrm{K}$. The inversion can be reversed by applying large positive bias to the junction. These observations are suggested to be inherent to Nb-$SrTiO_3$ when tunneling through the Schottky barrier occurs due to the non-linear ϵ_r of Nb-$SrTiO_3$ as this causes the Schottky barrier height and shape to change significantly with temperature.

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