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Summary

The realization of the transistor has been one of the most disruptive technological discoveries in the last century. Bloomberg Businessweek ranked it as the nr. 2 most disruptive idea in the last century saying: “The transistor […] and the ability to etch millions of them onto microchips became to the Digital Age what the steam engine had been to the Industrial Revolution.” Especially the extreme miniturization of the transistor size from tens of μms down to 10 nm presently. This miniturization has let to cheaper, faster and more energy efficient processors enabling appliances such as the personal computer, laptops and smart phones. Obviously the extremely successfull strategy of miniturization will hit a fundamentally obstacle: at some point it will reach the size of single atoms. It turns out that well before such the limit is reached the transistor already runs into trouble: the same minuturization, which led to better peformance, causes trouble at the very small scale: further downscaling increases power consumption and posses difficulties with heat dissipation. Additionaly, the demand to process very large volumes of data extremely fast is growing. This is problematic in such CMOS based technology because logic and memory are relatively far removed from eachother which causes CPU stalling. It is clear that technology based on a different principle is needed to realize long-term improvement, this technology is generally reffered to as beyond-CMOS.

A large class of beyond-CMOS devices are investigated which will most likely supplement (rather than fully replace) CMOS logic. Spintronic devices constitute a subset of possible beyond-CMOS device concepts which utilize in one way or another the quantum mechanical electron spin to realize memory or logic devices. There are many different spintronic device concepts which can be envisioned to perform logic operations. But before spintronic devices can be realized basic reseach is needed to investigate the feasibilty of these concepts. One very important building block is to realize spin injection into a semiconductor.

A few years before the start of my PhD project in 2011 several exciting advances had been made using a *three terminal geometry* to electrically create and detect spin

accumulation in p- and n-Si. In 2009 Dash et al. realized room temperature spin injection/detection in p- and n-doped Si. In 2011 they also proposed a convincing proposition to explain the presence of the unexpected in-plane magnetic field MR, dubbed the 'inverted-Hanle' effect. The observation of Seebeck spin-tunneling by Le Breton et al. provided even firmer grounds for the three terminal spin injection and detection method. Although spin signal amplitudes well above the theoretical maximum were observed Tran et al. had proposed a model which explained this by invoking spin accumulation in interface states. Much earlier the three terminal geometry was already used to show spin accumulation in n-GaAs by Lou et al. while detection in n-Ge was realized early 2012 by Kasahara et al. although both at low temperatures.

Inspired by this success this thesis aimes towards realizing and understanding spin injection in a semiconductor from an unorthodox class of materials: complex (transition-metal) oxides. Many of the different groundstates of matter can be found in complex oxides such as ferromagnetism, ferroelectricity, piezoelectricity, superconductivity et cetera. Hence, oxide based (spin)eletronic devices could benefit from the very versatile family of transition metal oxides and may open the way towards (spin)transistors with multiple degrees of freedom. This material class has already shown very intriguing properties for spintronic applications, but spin injection in a complex oxide semiconductor had not yet been reported. Semiconducting $SrTiO_3$ forms a natural choice since it serves as an epitaxial growth template for many complex oxides, is easily made semiconducting and shows an intriguingly large and strongly non-linear dielectric permittivity. Even more surprising a 2 dimensional electron gas (2DEG) can form at the $SrTiO_3$ surface when interfaced with other complex oxide insulators such as $LaAlO_3$, binary oxides such as γ-Al_2O_3 or at its bare surface by vacuum annealing. Despite the relatively low atomic mass of the constituent atoms large and electric field controllable Rasbha spin-orbit coupling had been shown.

This thesis starts out with an introductory chapter discussing the interest in spintronics, the current status of spin injection using a three terminal geometry and the current understanding and issues with such a geometry to probe spin accumulation.

In chapter 2 I introduce several concepts which are used throughout the thesis to interpret and understand the experimental work in the following chapters. I introduce the basic physics concerning the formation of a potential barrier when a metal and semiconductors are interfaced (referred to as a Schottky barrier). Additionally, I describe the different charge transport mechanisms with which electrons can traverse such a Schottky barrier. Since the presence of a Schottky barrier is in general detrimental to spin injection I also mention several ways a thin insulating layer can reduce the barrier height and width. Next I introduce the basic concepts related to spintronics such as the three-terminal spin detection technique, the formation of a spin accumulation, the influence of the ferromagnetic contact on the spin accumulation and discuss several ways that spin-obit coupling can influence the spin lifetime. Additionally, I introduce the concept of tunneling anisotropic magnetoresistance (TAMR) which also plays an important role throughout the thesis.

As mentioned, tuning of the interfacial electrostatics (for instance removal or reduction of the Schottky barrier) is important for realizing spin injection into a semiconductor. In chapter 3 we explore charge transport through ferromagnetic tunnel contacts on top of Nb:$SrTiO_3$. To tune the charge transport we insert a thin AlO_x layer in between the ferromagnet and Nb:$SrTiO_3$ and vary its thickness. The insertion of this ultra-thin AlO_x layer significantly suppresses the height and width of the tunnel barrier leading to a large increase in the junction conductance. Moreover, we are able to tune the charge transport from thermally assisted field emission to pure field emission when the AlO_x thickness is set at $\sim$1.1 nm while still maintaining a space-charge region at the semiconductor side. We developed an electrostatic model to describe the potential at a metal/insulator/n-$SrTiO_3$ interface which incorporates the temperature and electric field dependence of $SrTiO_3$'s permittivity. We show that the reduction in Schottky barrier height, due to the inserted AlO_x layer, is expected and consistent with this model. Our model also predicts very complex behavior of the electrostatic landscape when changing the temperature and voltage bias of the spin injection contacts. This has important implications for the charge and spin transport across these junctions as further discussed in chapters 4, 6 and 7.

In chapter 4 we demonstrate the realization of a spin accumulation at the interfacial region of Nb:$SrTiO_3$ with Co/AlO_x(11 Å) spin injection contacts at room temperature. We demonstrate a strong influence of the built-in electric field, close to the interface, on the spin lifetime in n-$SrTiO_3$ which varied from 2.5 to 17.5 ps. The manipulation of the built-in electric field, by biasing the junction, leads to a large change in the spin lifetime and its anisotropy. This is shown to be consistent with theoretical model calculations based on SOF spin flip scattering by manipulation of the Rashba SOC strength. A similar control over the spin lifetime and its anisotropy is shown by exploiting the electro-resistive control over the interface electric field. Using this electro-resistive effect the spin injection junction can be set to either a low or high resistance state which allows non-volatile control over the Rashba SOF strength.

Chapter 5 discusses the devices studied in the previous chapter in more detail. The influence of the fit function, with or without including spin diffusion, to extract the spin signal parameters is discussed. It is shown that the use of a fit function including spin diffusion results in larger spin lifetimes, large spin voltage and a larger ratio of the out- over in-plane spin voltage. However, the trends of these parameters as function of the junction voltage are the same for both fit functions. I also discuss the junction response when applying an in-plane magnetic field which reveals the presence of the 'inverted' Hanle effect. The direct extraction of the linewidth is problematic due to the presence of a high field linear MR and an anomalous low field MR effect. The bias dependence of the inverted Hanle amplitude shows correlation with the interpretation presented in chapter 4. I also discuss the influence of TAMR and show that when it is present the out-of-plane spin voltage can no longer be uniquely defined. In addition I argue that, for these devices at room temperature, the presence of TAMR can not be ruled out but must be small compared to the spin signal.

In the past years it has become increasingly more clear that it is difficult to determine what exactly is measured using a three terminal geometry. For instance, spin signal amplitudes many orders of magnitude larger than theoretically expected are observed in certain spin injection devices employing an oxide tunnel barrier. Generally these devices also show very similar spin lifetimes of around 150 ps. To explain this it has been proposed that instead of spin accumulation a novel MR effect is at the origin of the measured device response. This effect has been dubbed: impurity assisted tunneling magnetoresistance (iaTMR). A key element in iaTMR is the presence of an in-plane MR with linewidths equivalent to spin lifetimes $\sim$150 ps and the presence of charge transport via interface states (generally residing in the oxide tunnel barrier). In a literature review I show that junction which do not employ an oxide tunnel barrier, nor show in-plane MR but have spin lifetimes larger than 1 ns also exhibit spin signal amplitudes which deviate from theory by many orders of magnitude (both smaller and larger). This implies that such measurements can not readily be explained by either the iaTMR model or spin accumulation models.

In chapter 6 we have investigated the electro- and magnetoresistive response of the Co/Nb:$SrTiO_3$ interface. We show the coexistence of TAMR and electroresistive (ER) switching at room temperature. The maximum amplitude of the room temperature TAMR effect is around 1.7 %. This is significantly larger than the room temperature TAMR effects reported using more conventional counter electrodes such as Au, Pt, Ta or n-GaAs. Additionally, the bias at which the maximum TAMR is found (−75 mV) is substantially larger than for devices reported in the literature. At the same time these junctions exhibit ER switching effects with an on/off ratio close to 80 at low bias readout voltage. The TAMR effect is only negligibly influenced when performing ER switching operations allowing the simultaneous use of both spin and charge as a memory element. The large amplitude of the TAMR effect is attributed to the large permittivity of Nb:$SrTiO_3$ and possibly the d-band character of both Co and $SrTiO_3$. The ER switching most likely has its origin in the movement of ionic charge close to the interface altering the Schottky barrier profile. The current devices are believed to be far from optimal and leave significant room for optimization of both the TAMR and ER effect by altering the Schottky tunnel barrier, stoichiometry of Nb:$SrTiO_3$ and changing the ferromagnetic material.

The temperature dependent behavior of the device studied in chapter 4 and 5 as well as those with a 7 Å thick AlO_x barrier are investigated in chapter 7. The spin signal amplitude reduces by more than an order at low temperature. Additionally, the measured MR lineshape clearly indicates the emergence of TAMR for the junctions with an 11 Å barrier. Below $\sim$130 K the sign of the spin signal amplitude becomes negative in a certain bias window, indicating a spin accumulation of opposite polarization. The width of this bias window becomes increasingly larger with reducing temperature. At all temperatures a positive, conventional, sign of the spin signal is obtained at large positive bias. We observe a slight increase of the in-plane spin lifetime up to $\sim$35 ps at 4 K and 1.2 V junction bias. In junctions with the thinner 7 Å AlO_x barrier we observe similar behavior although a double sign change is observed. This behavior is observed for all junction studied. We propose that it is the inherent dependence of the Schottky barrier profile on temperature and bias that results in the bias dependent sign inversion at lower temperatures. This temperature dependence originates from the electric field in the space-charge region which leads to non-linearity of $Nb{:}SrTiO_3$'s permittivity. Since the Schottky barrier profile change is fully determined by the non-linearity of the permittivity a universal, temperature driven, inversion of the spin signal is expected as long as spins are injected through a space-charge region with large enough electric field. This mechanisms to tune the spin transport does not exists for linear dielectrics such as conventional semiconductors.