Charge and spin transport in Nb-doped SrTiO3 using Co/AlOx spin injection contacts
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Chapter 8

Conclusions
8. Conclusions

8.1 Conclusions, remarks and outlook

The work presented in this thesis was performed between the end of 2011 and mid 2016. In this period several different research groups reported spin injection experiments in Nb: SrTiO$_3$ or in the 2 dimensional electron gas residing in SrTiO$_3$ when interfaced with certain insulators. Additionally, it became apparent that the interpretation of three-terminal spin injection experiments is more complicated than previously realized. Here I will discuss the main results obtained in this thesis and discuss those in comparison with results obtained by other research groups. I will also discuss what my viewpoints are with respect to the interpretation of our measurements and the recently proposed impurity assisted tunneling magnetoresistance (iaTMR) effect.

The first step towards realizing spin injection into a semiconductor is by creating spin polarized tunnel contacts to the semiconductor. This has been extensively investigated for conventional semiconductors [1–4]. However, hardly any literature existed related to realizing such contacts to Nb: SrTiO$_3$. In chapter 3 such contacts are realized in the form of Co/AlO$_x$ heterostructures on Nb: SrTiO$_3$. Here it is shown that despite the much wider depletion region which forms at a Schottky contact with Nb: SrTiO$_3$, due to its large $\epsilon_r$, the same large $\epsilon_r$ causes a very rapid reduction of the Schottky barrier height and width when inserting a very thin layer ($\sim$1 nm) of AlO$_x$. This was shown to be qualitatively consistent with the electrostatic model we developed which takes into account the electric field and temperature dependence of the relative permittivity. In the same period very significant work using polar insulating interfacial layers for contact engineering was performed in Harold Hwang’s group [5–7]. Such flexible engineering of the interface potential shows great potential for device applications, where control over the interface is often crucial.

In April 2012 Reyren et al. reported spin accumulation in the 2DEG at the LaAlO$_3$/SrTiO$_3$ interface [8] at low temperatures. Roughly one year later Han et al. reported the first three terminal measurements indicating spin accumulation in Nb- and La-doped SrTiO$_3$ [9] at room temperature. In both reports the charge transport exhibited significant temperature dependence. Although not explicitly discussed in these works the $I–V$ data suggests that the junctions have a strongly temperature dependent zero bias conductance. These reports showed spin lifetimes of 50 (2DEG) and 75 ps (Nb: SrTiO$_3$) without a strong temperature dependence. Both reports showed spin-RA products many orders of magnitude larger than theoretically expected: 60 MÎ©µm$^2$ (2DEG) and 40 kÎ©µm$^2$. Additionally, they both reported the presence of the inverted Hanle effect with amplitudes much larger than the Hanle amplitude. In the case of Han et al. the supplementary material also showed that the Hanle and inverted Hanle signal did not exhibit any anisotropy at large fields. Note that both reports conclude that they do not measure the spin lifetime of the SrTiO$_3$ bulk. Reyren et al. invoke spin accumulation in interface states while Han et al. suggest that they measure an interface limited spin lifetime (due to formation of
magnetic Ti$^{3+}$ centers at the MgO/n-SrTiO$_3$ interface).

The results presented in chapter 4 are in stark contrast with those in the previous paragraph. The charge transport is weakly temperature dependent (expected for tunneling) and the zero bias conductance decreases by less than a factor 2 at 4 K. An analysis of the temperature dependent saturation current indicates that tunneling occurs through the oxide and Schottky tunnel barrier. The presence of the Schottky barrier is also indicated by the increased transparency at reverse bias as discussed in chapter 3. The measured in-plane spin lifetime $\tau_{||}$ systematically changed from 2.5 to 17.5 ps at room temperature, overall much lower than in the previous studies. More importantly both $\tau_{||}$ and the ratio of the out-of-plane to in-plane spin lifetime $\tau_{\perp}/\tau_{||}$ show a systematic increase with increasing forward bias voltage, and hence reducing interface electric field strength. It was shown that this behavior is consistent with changes in the spin flip scattering by modulating the strength of a Rashba-like Spin-Orbit-Field (SOF).

Note that this implies that Rashba SOC strongly limits the spin lifetime at the injection/detection contact when a large enough electric field is present. Hence this work suggests that proper care has to be taken to significantly suppress any electric-fields at these interfaces. On the other hand, the reduction of the spin lifetime using electric field can be useful in certain spin transistor concepts [10]. The maximum spin lifetime observed is $\sim$17.5 ps which leads to a spin relaxation length $\lambda$ just below 20 nm (using $D_s = 0.2 \text{cm}^2 \text{V}^{-1} \text{s}^{-1}$). Theoretical calculations show that such a short spin lifetime is not expected, intrinsically, for n-SrTiO$_3$ [11]. However, in addition to Rashba SOC, SrTiO$_3$ crystals contain many defects and (magnetic) impurities, even when undoped, which will reduce the spin lifetime from those theoretically calculated. The analysis in chapter 5 suggests that random magneto-static fields limit the spin lifetime at large forward bias. Hence it seems likely that much longer spin lifetimes can be obtained in higher purity, defect free n-doped SrTiO$_3$ or inside the 2DEG which form at the surface of SrTiO$_3$ interfaced with numerous insulators.

Also note that the band structure of SrTiO$_3$ is relatively complex, especially when degeneracy is lifted by electric fields, strain, off-stoichiometry et cetera. This makes it unclear which band(s) the spins populate and hence complicate the extraction of the spin lifetime. For instance, the effective mass $m^*$ could be significantly larger than the free electron mass $m_e$ (reports of up to $10m_e$ exist [12]), a large $m^*$ would have resulted in an underestimation of the extracted $\tau_{||}$. Assuming $m^* = 10m_e$ this would lead to an increased $\lambda$ of around 60 nm at room temperature. Using a four-terminal non-local detection scheme would still be practically impossible for such a short $\lambda$.

In chapter 4 we also show that the electro-resistive manipulation of the junction resistance can be used to alter the spin signal amplitude and spin lifetime. The specifics of the manipulation are also consistent with the proposed electric field controlled Rashba SOF spin flip process. This indicates that it is not only possible to control the spin lifetime electrically but also in a non-volatile way. Such additional functionalities
are interesting for the development of energy efficient beyond-CMOS technology.

We observe a strong reduction in the spin signal amplitude with reducing temperature as discussed in chapter 7. At \( T \lesssim 130 \) K the sign of the spin signal becomes bias dependent. The observation of a Hanle dip instead of a Hanle peak suggests the polarization of the accumulated spins has reversed. At larger forward bias the sign reverts to its conventional positive value (Hanle peak). There have been several reports of such a sign reversal, mostly for epitaxial Fe contacts on n-GaAs, at low temperatures. In those works the reversal is often attributed to the presence of interface resonance states. In our work the spin injection contacts are not epitaxial, see chapter 3, making sign inversion via resonance states unlikely. In addition we observe similar behavior of the spin signal amplitudes for many different junctions. In contrast, the results for the Fe/n-GaAs junctions widely vary from device to device probably related to the detailed interfacial structure. We propose that this is a unique feature of SrTiO\(_3\)’s large and strongly non-linear \( \epsilon_r(E,T) \) causing significant changes in the interfacial potential profile forming the tunnel barrier. Since it has been shown that the shape change of a tunnel barrier can lead to such a bias dependent sign inversion this seems a natural explanation. Currently theoretical calculation are ongoing to substantiate this claim.

The maximum in-plane spin lifetime increases by roughly a factor 2 (~35 ps) at 4 K at 1.2 V. This weak increase is in contrast with the large increase of the electron mobility by almost 2 orders of magnitude (from \( \sim 10 \) to \( \sim 10^3 \) cm\(^2\) V\(^{-1}\) s\(^{-1}\)). This again suggests the the spin lifetime is limited by some other scattering mechanism. Note that the increased mobility is correlated to the increased electrostatic screening at low temperatures due to the large \( \epsilon_r \). Hence, close to the interface, where \( \epsilon_r \) deviates from the bulk value, the mobility could be much lower than in the bulk. It is also possible that the applied bias is large enough to create an accumulation layer at the interface which could be expected to alter the spin lifetime. Note that the increased spin lifetime and large increase in electron mobility should greatly increase the spin relaxation length. Assuming a spin lifetime of 37 ps and a 100 times increased \( D_s \) a spin relaxation length of around 300 nm is obtained which is in principle possible to detect using a non-local geometry.

These observations are again in stark contrast to those reported by Ref. [8] and [9] since neither observe a sign inversion. Normally a weakly increasing [9] or strongly increasing [13] trend of the spin signal amplitude is observed with reducing temperature in almost all three terminal spin injection measurements in semiconductors (including those in SrTiO\(_3\)). Both the SrTiO\(_3\) based works did not observe a temperature dependence of the spin lifetime. The absence of the complicated behavior as observed in our work is consistent with the fact that they employ thick tunnel barriers which fully dominate the junction resistance. Hence, the physics associated with the depletion region in n-doped SrTiO\(_3\) should not be observed.

A downside to studying spin injection in a three-terminal geometry is its local na-
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ture. Firstly, other spurious magneto-resistance effects can arise due to the local nature of the three-terminal method. At low temperatures tunneling anisotropic magneto-resistance (TAMR) is clearly present in the devices with 11 Å thick tunnel barriers. In chapters 5 and 7 we show that this restricts the information that can be extracted from the Hanle measurements. Nonetheless, we are still able to obtain information about the in-plane spin lifetime and voltage as shown in chapter 7.

The presence of TAMR in itself is however of great interest. In chapter 6 we show that Co/Nb:SiTiO$_3$ Schottky interfaces exhibit a large room temperature TAMR effect along with electroresistive (ER) switching. The TAMR effect is roughly 5 times larger than those obtained in previous works not employing doped SrTiO$_3$ [14–16]. Significant improvements of the TAMR effect size seem feasible as we believe that the charge transport is not yet optimal to obtain the maximum TAMR effect. Also note that the use of Co, which has a relatively weak spin-orbit coupling, is not expected to be optimum leaving room for further improvements. Combining the TAMR with the large electroresistive effects at these contacts might allow to store multiple logic states. One could even envision device schemes where the ER or TAMR effect could store bits while serving as a spin injection contact for a spin based transistor, leading to extremely close integration of memory and logic functions. Note that closing the gap between memory and logic is expected to lead to strongly improved performance for instance by reducing the energy-delay product [17].

It has been shown that it is complicated to distinguish if the observed Lorentzian MR originates from dephasing of a spin accumulation or other effects such as iaTMR [18, 19]. In the work presented in this thesis we observe several features which are consistent with those expected for the iaTMR effect. We observe the ‘inverted’ Hanle effect and obtain spin signals which are considerably larger than those expected for spin accumulation in Nb:SiTiO$_3$ bulk in the linear regime (up to roughly 10 times to larger when using $P = 1$). However, we also observe many features which are not readily explainable within this theoretical framework. The main ones are:

- **The overall broader linewidth compared to Refs. [18–20], also compared to the studies in Nb:SiTiO$_3$ [7, 9].** This would imply a much larger effective local magnetic field $B_L$. We have no indications $B_L$ should strongly differ from those present in the works of Refs. [7–9].

- **The systematic change in linewidth (i.e. $\tau_\parallel$) with bias.** This would require a systematic change in $B_L$ as function of junction bias, which we believe is unlikely but cannot be ruled out.

- **The systematic change of $V_\perp/V_\parallel$ ratio with bias.** Although in the iaTMR framework no anisotropy is expected, another magnetoresistance effect (e.g. TAMR) could cause such an anisotropy if it has the correct bias dependence. We have shown TAMR to be present (see chapter 6 and at low temperature in chapter 7) and its...
amplitude is shown the decrease with forward bias. However, as discussed in chapter 5, the reverse bias measurements do not indicate a large TAMR effect to be present at room temperature for the devices with 11 Å AlO$_x$.

- **Measurements in the LRS and HRS state show consistent values of $\tau_\parallel$ and $V_{\perp}/V_{\parallel}$ as function of junction voltage.** The fact that the junction exhibit electroresistive effects strongly indicates defects to be present at the interface. If these defects play a direct role in creating the spin signal (i.e. involved in the iaTMR effect, or spin accumulation in interface states) it is unlikely that the junction response would be the same (after changing their spatial position or occupation) at the same voltage bias. The observed behavior is, however, consistent with the proposed spin dephasing via Rashba SOC.

- **The spin signal amplitude strongly decreases with reducing temperature.** The iaTMR effect should exhibit a strongly increasing amplitude with reducing temperature (as discussed in Ref. [18, 19]).

- **The sign of the spin signal is bias and temperature dependent.** Inversion of the sign would be possible within the iaTMR theory if the magnetization anisotropy of the cobalt layer is altered such that its easy axis is rotated to the out-of-plane direction. This does not happen in our devices. I would like to note that if the spin transmission of the Schottky barrier is indeed altered with bias, as suggested in chapter 7, it might be possible to obtain a sign inversion within the iaTMR framework. This does imply that the relevant defect states do not lie inside the oxide barrier or at the interface between the oxide barrier and the semiconductor but states inside the depletion region. Therefore this would not change the main message of chapter 7 which discusses tuning of the spin transmissivity of the Schottky barrier in Nb:SrTiO$_3$ diodes.

Within the framework of spin accumulation, the presence of the ‘inverted’ Hanle effect implies that we are not probing the intrinsic spin lifetime of Nb:SrTiO$_3$. Additionally, the large amplitude of the spin signal indicates a possible enhancement of the spin detection sensitivity of the contact. While the enhanced spin signal amplitude can in principle originate from spin accumulation in interface states this is inconsistent with the Rashba SOC interpretation (since interface states are non-propagating). In the light of all the complexity concerning interpretation of the three-terminal spin injection measurements I feel it is not possible to claim spin accumulation in the bulk semiconductor. However, the data strongly suggests that Rashba SOC is influencing the spin lifetime. The rich behavior of the spin signal amplitude and sign at low temperatures also provide strong evidence that spin transmission through the Nb:SrTiO$_3$ interface is observed, regardless of the exact details. The unique bias and temperature dependence of the spin signals, large TAMR and ER, very different from
those observed in three-terminal spin injection studies of conventional semiconductors, show that n-SrTiO$_3$ forms a fruitful ground for exploring (spin)electronic effects and devices.

### 8.2 Outlook

One of the first issues which needs to be resolved is the understanding and interpretation of three-terminal Hanle measurements. Most importantly a method is needed to separate the observed magnetoresistance response from the iaTMR effect, spin accumulation in interface states and spin accumulation in proximity of the contact. Recently, there has been significant progress on this front. Inokuchi et al. have shown that it might be possible to separate out the contribution of defect states related magnetoresistance from actual spin accumulation under the injection contact [21]. They argue that with the combined use of DC and AC biasing techniques the magnetoresistance effect related to transport via defects can be filtered out. The idea is that at a large enough AC measurement frequency the voltage change across the electrode is fast enough to prevent intermediate states from participating in conduction, hence preventing those states to play a role in the magnetoresistance. This would in principle allow to map out the component related to defect states and to bulk spin accumulation by performing frequency dependent magnetoresistance measurements.

It has also become clear that the extraction of the spin lifetime in four-terminal non-local Hanle measurements can be unreliable. This is mainly the case when the distance $d$ between the injection and detection contact becomes small (i.e. below $\lambda$) and the contact are relatively transparent (hence the ferromagnets acts as spin sinks) [22][24]. A three-terminal geometry can be viewed as a four-terminal geometry where the $d \to 0$ and hence should be extremely susceptible to influence from the ferromagnetic contacts. Additionally, the contact area in three-terminal measurements is generally much larger than the spin diffusion length. Any spatial fluctuations in the tunnel barrier thickness will result in local patches under the contact where the ferromagnetic contact is more or less invasive. This will influence the local spin accumulation size and effective spin lifetime. It might be that such inhomogeneities play a role in the measured spin signals which are currently not accounted for. To incorporate this, 2D or 3D spin accumulation models might prove to be important for the interpretation of three-terminal Hanle measurements. Along with other issues, such as stray fields, it remains difficult to understand in how far a three-terminal geometry probes the intrinsic bulk spin lifetime of a non-magnetic material.

It seems thus vital that three-terminal measurements are followed up with methods that probe the transport in the channel more directly. Although the spin lifetime obtained for Nb: SrTiO$_3$ in this thesis suggests that the spin diffusion length is only several tens of nanometers at room temperature, it seems strongly limited by inter-
face properties and the low purity of the crystal. Using low defect thin films of doped SrTiO$_3$ or one of the many 2DEGs can be expected to yield much larger spin diffusion lengths allowing four-terminal measurement schemes. It would be of great interest to investigate if, in such channels, electric field tunable Rashba SOFs allow similar manipulation of the spin lifetime but over more orders of magnitude. Additionally, more advanced control over the growth of doped SrTiO$_3$ should allow the creation of a thin δ-doped surface layer. This could open the pathway towards spin injection in much lower doped SrTiO$_3$ and remove the need for interfacial oxide tunnel barriers. At the same time this will create a spin injection interface with a large local electric field which could allow electric field control over the spin species that is injected in the channel, although this might be paired with modulation of the Rashba SOC strength. Increased control of the interfacial electrostatics might also allow enhancing the large room temperature tunneling anisotropic magnetoresistance effects present at Co/Nb: SrTiO$_3$ junctions while retaining the electroresistive switching.

Additionally one might envision epitaxial integration of other complex oxides with exciting properties such as piezoelectricity and magnetoelectricity or even multiferroic properties. Such elements are of significant interest for beyond CMOS technology as it would allow electric field control over the magnetization state.

One of the key findings of this thesis is that spin injection contacts on Nb: SrTiO$_3$ exhibits different, often more rich, spin physics than observed for conventional ferromagnets. This, at the very least, makes SrTiO$_3$ an interesting playground to explore spintronic effects but also holds promise for the realization of spintronic devices.

References
