Spin transport in normal and superconducting nanowires

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Abstract

Today's conventional electronic devices are based on electron charge transport in semiconductor channels. Spintronics is a rapidly emerging technology, which exploits the spin degree of freedom as well as the charge of the electrons. It is believed that extending conventional electronics to spin-electronics can yield devices with new functionality and result in new large scale applications. Examples of already existing spintronic technology are the magnetic random access memory, magneto-resistive read heads in hard drives and various magnetic field sensors. The fundamental requirement for a working spintronic device is the ability to generate, transport and detect spin currents, which are the subject of this thesis.

A current, spin polarized by a ferromagnet and injected into a non-magnetic material remains polarized for the duration of the spin relaxation time. This relaxation time, and consequently the useful distance the injected non-equilibrium spin can be transported in the non-magnetic transport channel, is dependent on the underlying spin relaxation mechanisms in the material. Furthermore, the transport channel can be devised to exploit the spin-orbit scattering within the channel with the aim to achieve novel spin transport effects, such as the Spin Hall effect. We study such mechanisms and effects in normal and superconducting nanowires. The main results of the work are the following:

In thin film devices, the thickness of the electron transport channel can be comparable to the electron's mean free path, which makes the surface scattering the dominant scattering mechanism. To investigate how the additional surface momentum scattering affects spin relaxation, the thickness dependence of the spin relaxation parameters was analyzed. Using spin injection into Al nanowires of various thickness, it was found that the spin flip scattering at the surfaces is substantially weaker compared to that within the bulk of Al.

A five terminal device having a pair of spin sensitive detector electrodes placed symmetrically about the injection point was used to directly demonstrate the decoupling of spin and charge currents in a one-dimensional transport channel. The spin accumulation is shown to be strictly symmetric about the injection point and independent of the direction of the charge current.

For superconducting nanowires, it is found that the spin accumulation is enhanced by up to 5 orders of magnitude compared to that in the normal state of the wire. In contrast, the spin diffusion length is found to decrease by an order of magnitude on transition in to the superconducting state. This is interpreted as due to magnetic impurity rather than spin-orbit dominated spin-flip scattering in the nanowires studied. We additionally observe a giant spin Hall effect in superconductors, which is more than 5 orders of magnitude stronger than the values reported recently for Al nanowires in the normal state.
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Chapter 1

Introduction

The electron was discovered in 1897 by J.J. Thomson, who was studying cathode rays. Approximately ten years later, R. Millikan measured the charge of the electron in his famous oil-drop experiment. It was not until 1922 that Stern and Gerlach discovered the magnetic moment of the electron. It was later established that the intrinsic angular momentum of the electron, or spin, is quantized such that, whenever it was measured it would yield either spin-up ($+\hbar/2$) or spin-down ($-\hbar/2$) projections. Today's conventional electronics does not use the spin of the electrons and relies only on the transport of the electron charge. Physicists are now trying to exploit the electron spin in order to create novel devices with a broader functionality. This rapidly emerging field is referred to as Spintronics.

The first observation of spin affecting electron transport dates back to 1857, when W. Thomson found that the resistivity of bulk ferromagnetic metals depended on the relative angle between the electric current and the magnetization direction [1]. This phenomenon is called anisotropic magnetoresistance (AMR) and has been used in e.g sensors for magnetic recording. Approximately a century later, Tedrow and Meservey performed the first study on spin polarized tunneling on ferromagnetic/insulator/superconducting aluminum junctions [2]. By applying a magnetic field along the film surface, the Zeeman effect makes it possible to determine the spin polarization of the tunneling current.

In 1975, Julliere reported an increase in resistance (∼10% at low temperature) when the magnetic layers in Fe/Ge/Co switched from the parallel (P) to the antiparallel (AP) configuration [3]. This phenomenon is called tunneling magnetoresistance (TMR) and is much stronger than the AMR effect. It can be explained by a simple analogy to a light polarization, in which un-polarized light will be blocked or passed through two polarizing plates if the polarizing axis are crossed or aligned, respectively. Similarly an un-polarized electron current will experience either a high (AP) or low (P) resistance, when passing through the two ferromagnetic layers in a magnetic tunnel junction, which act as spin polarizers for electrons. Twenty years later Moodera et. al. were able to observe a TMR of
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about 20% at room temperature, the improvement being due to optimized junction preparation procedures and the choice of ferromagnetic materials [4]. A third well known magnetoresistance effect is the giant magnetoresistance (GMR) effect, which was discovered in 1988 [5–7]. Like the TMR elements, GMR devices consist normally of two ferromagnets separated by a non-magnetic metal. GMR and more recently TMR elements have enabled significant improvements in hard drive read heads, magnetic sensors and memory chips. Nanomagnets can be used to store information without the need for a power supply. Such a non-volatile memory is known as a magnetic random access memory (MRAM), and has recently become commercialized.

The fundamental requirement for a working spintronic device is the ability to create, transport and detect spin currents. The idea to create a spin population in a non-magnetic metal, by driving a current from a ferromagnet into the non-magnet, was proposed by Aronov and Pikus already in 1976 [8]. It was not until ten years later when Johnson and Silsbee, in their pioneering experiments, first demonstrated electrical spin injection, accumulation and detection in single crystal bulk aluminum at low temperatures [9–11]. They used a ferromagnetic electrode to inject spin into the Al and another ferromagnetic electrode, placed outside the charge current path, to measure the non-equilibrium spin accumulation. This non-local measurement technique has become widely used and helped to uncovered a number of new spin-dependent phenomenon in nanostructures [12–19]. The strength of this experimental method is that it allows to directly measure non-equilibrium spin accumulation and relaxation. Direct, electrical read-out observations of electron spin accumulation and precession [12, 13], spin Hall Effects [16, 19], and spin injection and propagation in Si [18] are some of the prominent examples in this development.

The work described in this thesis focuses on the injection, relaxation and detection of spin currents in one-dimensional transport channels. The spin transport properties are investigated in both the normal and superconducting state. The outline of the thesis is the following:

In Chapter 2, the resistivity of films is discussed in terms of the main scattering mechanisms. A special attention is paid to surface scattering in thin films.

In Chapter 3 the spin dependent transport is discussed in detail. Main issues are how to effectively create, transport and detect spin. Also the mechanisms of spin relaxation are discussed.

Chapter 4 is an introduction to superconductivity in relation to spin dependent transport.
In Chapter 5 the spin Hall effect in both the normal and superconducting states is discussed.

Chapter 6 describes the sample fabrication process and the measurement techniques.

Chapter 7 illustrates the main experimental results on spin transport in nanowires in the normal state.

Chapter 8 discusses the measurements on spin transport in the superconducting state.

Chapter 9 describes the results of the spin Hall effect experiments in the superconducting state.
Chapter 2

Thin Film Resistivity

2.1 Introduction

One of the first laws a physics student gets to learn is Ohm’s law, which states that passing a current \( I \) through a conductor generates a voltage drop \( V \) that is proportional to the resistance \( R \) of the conductor, \( V = RI \). The resistance depends on the resistivity, \( \rho \), of the material according to

\[
R = \frac{\rho L}{A},
\]

(2.1)

where \( L \) is the length and \( A \) is the cross-sectional area of the material. Normally, \( \rho \) is constant and does not depend on the geometry of the sample. It is a measure of the impurity level in the material. This, however, is only true for bulk materials or thick films, where the thickness \( d \) is much larger than the mean free path \( \lambda_0 \) of the conduction electrons, i.e. the majority of the scattering occurs inside the bulk material. As the thickness becomes comparable to or smaller than the mean free path the resistivity increases rapidly. This enhancement is often attributed to the influence of surface scattering and in the case of granular films also grain boundary scattering. According to the Matthiesen rule, these contributions are additive

\[
\rho_{\text{tot}} = \rho_{\text{bulk}} + \rho_{\text{surface}} + \rho_{\text{grain}}
\]

and can be modeled separately.

2.2 Theoretical Models

The interest in this subject has been substantial for decades and a lot of theoretical [20–30] and experimental [31–42] studies has been made to achieve a better understanding. The different theoretical approaches have been considered and compared to the experiment in order to determine the main scattering mechanism in thin films. Throughout this section we assume that the temperature is low, such that phonon scattering is negligible.
Fuchs-Sondheimer model

Fuchs in 1938 and Sondheimer in 1952 [20,21] developed a theory for surface scattering of electrons in parallel-sided metal samples, which has often been utilized in this field. It is particularly attractive because it depends only upon two parameters, these being the ratio between the thickness and the bulk mean free path, \( \kappa = \frac{d}{\lambda_0} \), and specularity parameter, \( p \), being the probability that an electron incident at the surface will scatter specularly. Fuchs and Sondheimer use the Boltzmann equation for free electrons to obtain the thin film resistivity, \( \rho_f \):

\[
\frac{\rho_0}{\rho_f} = 1 - \frac{3}{2\kappa} \int_0^1 du \left( u - u^3 \right) \frac{(1 - p)(1 - \exp(-\kappa/u))}{1 - p \exp(-\kappa/u)},
\]

where \( \rho_f \) and \( \rho_0 \) are the film and bulk resistivity, respectively. Although widely used, this theory has fundamental limitations. These are that the Fermi surface is spherical, the mean free path is isotropic and the surface scattering is independent of the incident angle. It also assumes that the films are single crystalline, i.e. no grain boundary scattering is present.

Soffer model

The idea of an angle dependent specularity parameter \( p(\theta) \) was introduced by Soffer [22]:

\[
p(\theta) = \exp \left\{ - \left( \frac{4\pi h}{\lambda_e} \right) \cos^2 \theta \right\},
\]

where \( \lambda_e \) is the electron wavelength, \( h \) is the r.m.s surface roughness and \( \theta \) is the incident angle. This formula is used in different fields of physics and is equivalent to those used in optical scattering models. Soffer obtained the same expression for \( \rho_f \) as Fuchs-Sondheimer except that the constant \( p \) is replaced with \( p(\theta) \)

\[
\frac{\rho_0}{\rho_f} = 1 - \frac{3}{2\kappa} \int_0^1 du \left( u - u^3 \right) \frac{(1 - p(u))(1 - \exp(-\kappa/u))}{1 - p(u) \exp(-\kappa/u)},
\]

This model can be extended to include two different \( p \)'s for the top and bottom surface.

Mayadas-Shatzkes model

The models described so far have ignored the effects of grain boundaries, always present in poly-crystalline films. Mayadas and Shatzkes [23] modeled the contribution from grain boundary scattering to the resistivity as follows:

\[
\frac{\rho_0}{\rho_f} = 1 - \frac{3}{2} \alpha + 3\alpha^2 - 3\alpha^2 \ln(1 + 1/\alpha) = G(\alpha),
\]
where
\[ \alpha = \frac{R \lambda_0}{1 - R D}, \]

\( R \) is the grain boundary reflection coefficient and \( D \) the average grain width. Combining this result with the Fuchs-Sondheimer theory they obtain the following expression for the resistivity due to surface and grain boundary scattering:

\[
\frac{\rho_0}{\rho_f} = G(\alpha) - \frac{6(1-p)}{\pi \kappa} \int_{0}^{\pi/2} d\phi \times
\]

\[
\times \int_{0}^{1} du \cos^2 \phi \left( \frac{(u - u^3)(1 - \exp(-\kappa H))}{1 - p \exp(-\kappa H)} \right),
\]

(2.6)

where \( H = 1 + \alpha \left\{ \sqrt{(1-u^2)\cos^2 \phi} \right\}^{-1} \). Now, scattering from the surface and grain boundaries is incorporated in one formula.

**Soffer-Mayadas-Shatzkes model**

The final step is to combine the Mayadas-Shatzkes model with the Soffer model in the case of two different surface roughnesses. This was done by Sambles et al. [27] who assumed angularly dependent surface specularities \( p_i(\cos \theta) \) and columnar grain growth of diameter \( D \) with specularity \( R \):

\[
\frac{\rho_0}{\rho_f} = G(\alpha) - \frac{4}{\pi} \int_{0}^{\pi} d\phi \int_{0}^{1} du \cos^2 \phi 3(u - u^3) \times
\]

\[
\times \frac{1 - \exp(-\kappa H/u)}{2\kappa H^2} \frac{\{1 - \bar{p} + (\bar{p} - p_1p_2) \exp(-\kappa H/u)\}}{\{1 - p_1p_2 \exp(-2\kappa H/u)\}},
\]

(2.7)

where \( \bar{p} = \frac{1}{2}(p_1 + p_2) \). This model is best suited for our studies since it includes all the necessary scattering contributions. In fact, it will be shown that it fits our experimental data very well and that the dominant scattering mechanism in our films is due to surfaces.
Chapter 3

Spin Dependent Transport

3.1 Ferromagnetism (F)

It is well known that a current in a loop produces a magnetic moment, $\mu = IA$, where $I$ is the current and $A$ is the enclosed area. The current consists of circulating charge $q$, $I = q/t$, where $t$ is the period. The direction of such moment is perpendicular to the plane of the loop, given by the right hand rule. In the same way, an electron orbiting around its nuclei with a period $t$ will produce an "orbital" angular momentum $L = 2mA/t$, which is proportional and parallel to its magnetic moment, $\mu = \frac{q}{2m}L$. Moreover, since the electron has an intrinsic "spin" angular momentum $S$, the total magnetization of the circulating electron is

$$\mu = -\mu_B(g_LL + gS),$$

where $\mu_B = 57.9 \mu eV/T$ is the Bohr magneton and the $g$-factors are $gL = 1$ and $g = 2$. The magnetic moment of protons and neutrons is about 2000 times smaller than $\mu_B$ which is why the magnetization from the nuclei is often neglected. Each electron can be considered as an extremely small magnet, but aligning many together will result in a sizeable macroscopic magnetic field.

The characteristics of ferromagnetism is that the number of spin-up and spin-down electrons in the material is unequal. This is true in ferromagnets (F) such as Co, Fe and Ni where the density of states (DOS) for the two spin sub-bands is shifted by the exchange energy. The exchange interaction is due to the overlap of the neighboring atomic orbitals, which can lead to spin-dependent interaction between the atomic magnetic moments. The electron wave function has to obey the Pauli exclusion principle: the total wave function of a two electron system has to be antisymmetric with respect to the interchange of any two electrons. It is the difference between the symmetric and antisymmetric spin part of the wave function that is referred to as the exchange energy. If this exchange energy is larger than the thermal energy, all magnetic moments are aligned, producing a large macroscopic moment.
Figure 3.1: (a) Simplified band structure of a ferromagnet. (b) Density of states (DOS) for bcc Cobalt, after [43].

Figure 3.1 illustrates two types of DOS, i.e the s- and d-character. The s-electrons are regarded as free (conduction) electrons with non-localized wave functions, whereas the d-electrons have a large effective mass making them relatively immobile and localized. The s-electrons scatter and couple to the d-electrons and thus get polarized. Seen from another perspective, the d-band for spin-up electrons is filled while for the spin-down electrons it is partly filled at the Fermi level. Therefore, the only available states are those for the spin-down carriers, which in turn leads to spin polarization of the conduction electrons.

3.2 Spin Polarization

Light is polarized when passing through an anisotropic optical material. In analogy, an electron current will be polarized when sending it through a F. Due to the shift in the DOS the spin-up (↑) and spin-down (↓) currents will have different conductivities given by the Einstein relation,

\[ \sigma_{\uparrow,\downarrow} = e^2 N_{\uparrow,\downarrow} D_{\uparrow,\downarrow}. \]

(3.1)

Here \( N_{\uparrow,\downarrow} \) is the DOS at the Fermi energy, \( D_{\uparrow,\downarrow} = 1/3v_{F\uparrow,\downarrow}l_{\uparrow,\downarrow} \) the diffusion constant, \( \sigma_{\uparrow,\downarrow} \) the conductivity, \( v_{F\uparrow,\downarrow} \) the average Fermi velocity and \( l_{\uparrow,\downarrow} \) the mean free path. The current can be considered to consist of two separate spin channels [44–46] where the sum corresponds to the charge current \( I_c = I_\uparrow + I_\downarrow \) and the difference corresponds the net spin current \( I_s = I_\uparrow - I_\downarrow \). By definition, the bulk current polarization is given by the ratio between the two currents, i.e \( P = I_s/I_c \). Since the currents are proportional to \( \sigma \) we obtain the bulk polarization in F as

\[ P_F = \frac{\sigma_\uparrow - \sigma_\downarrow}{\sigma_\uparrow + \sigma_\downarrow} = \frac{N_\uparrow - N_\downarrow}{N_\uparrow + N_\downarrow}. \]

(3.2)
3.3. TUNNELING SPIN POLARIZATION

In this thesis, we define the spin-up band to be related to the majority electrons, which determines the magnetization direction, and the spin-down band to be related to the minority electrons. This is a common definition in the field.

Determining the magnitude and sign of $P$ is not a trivial task. One has to consider spin dependent scattering of the conduction electrons in $F$ and not only the electronic band structure. In the case of Co, one would expect a negative $P_F$ since the d-band for the spin-up electrons is filled (below $E_F$) whereas it is partly filled for the spin-down electrons. However, Eq. 3.2 does not take into account the complexity of the Fermi surfaces and the fact that s-electrons have higher Fermi velocities and thus contribute more to the current than the d-electrons. A more accurate definition is given by Mazin [47]

$$P_n = \frac{\langle Nv^n \rangle_\uparrow - \langle Nv^n \rangle_\downarrow}{\langle Nv^n \rangle_\uparrow + \langle Nv^n \rangle_\downarrow}, \tag{3.3}$$

where

$$\langle Nv^n \rangle_\sigma = (2\pi)^{-3} \sum_\alpha \int v^\alpha_{k}\delta(E_{k}\sigma) d^3 k, \quad \sigma = \uparrow, \downarrow, \tag{3.4}$$

$E_k$ is the energy of an electron in band $\alpha$, with wave vector $k$, $v^\alpha_{k}\delta$ the velocity and $n$ is an index depending on the kind of experiment. Tsymbal et al. [48] made ab initio band structure calculations that take into account the spin independent and spin dependent scattering and predicted a positive $P_F$ for Co of about 60%. There are several ways of both experimentally and theoretically obtaining the polarization.

3.3 Tunneling Spin Polarization

When a $F$ is in contact with a tunnel barrier, spin polarization (SP) is the polarization of the tunneling electrons and is not an intrinsic property of the $F$ alone. It depends on the structural and electronic properties of the entire junction including the interfaces. Unlike Eqs. 3.2-3.3, which gives the bulk polarization in $F$, it is therefore difficult to find a general theoretical model for the tunneling polarization, $P_T$. The standard definition of $P_T$ is

$$P_T = \frac{G^\uparrow - G^\downarrow}{G^\uparrow + G^\downarrow}, \tag{3.5}$$

where $G^\uparrow$ and $G^\downarrow$ are the tunnel barrier conductivities of the spin-up and spin-down channels, respectively.

In the 1970’s Meservey and Tedrow [2,49–51] developed a technique to measure the amplitude and sign of the SP for various $F$’s in their pioneering tunneling experiments on Ferromagnet/Insulator/Superconductor (F/I/S) junctions. The superconductor used as a spin detector was Aluminum. When a magnetic field $H$ is applied parallel to the film plane, the quasiparticle DOS in the S are split by
2μ_b H due to the Zeeman interaction into the spin-up and spin-down populations, see Figure 3.2 (a). Due to unequal DOS at the Fermi energy in the F the tunneling conductance into S becomes asymmetric, see Figure 3.2 (b). Assuming that the tunneling is spin conserving, the spin polarization $P_T$ is given by the relative heights of the four conductance peaks $\sigma_{1-4}$ as:

$$P_T = \frac{\sigma_4 - \sigma_2 - (\sigma_1 - \sigma_3)}{(\sigma_4 - \sigma_2) + (\sigma_1 - \sigma_3)}.$$

Taken into account spin-orbit scattering in the S enables a more accurate determination of $P_T$. Using this method Meservey and Tedrow determined the polarization for various F's, $P_{Fe} = 44\%$, $P_{Co} = 34\%$, $P_{Ni} = 11\%$ and $P_{Gd} = 4.5\%$.

**The Stearns model [52]**

Stearns developed a simple model to treat SP in various F metals. She pointed out that the transmission probability depends on the effective mass which is different for different bands. As mentioned earlier, the d-electrons are localized and have large effective mass and therefore decay very rapidly into the barrier, whereas the s-electrons are light and decay slowly. Accordingly, the nearly free electron bands should provide essentially all the tunneling current. Assuming parabolic s-bands, $E \propto k^2$, the DOS of these bands at the Fermi energy are proportional to their Fermi
wave vectors, i.e. \( N_\uparrow \propto k_\uparrow \) and \( N_\downarrow \propto k_\downarrow \). Eq. 3.2 then reads

\[
P_F = \frac{k_\uparrow - k_\downarrow}{k_\uparrow + k_\downarrow},
\]

where \( k_\uparrow \) and \( k_\downarrow \) are the Fermi wave vectors of the spin-up and spin-down bands. Stearn found that \( P_{Fe} = 45\% \) and \( P_{Ni} = 10\% \), which are consistent with the values determined by Meservey and Tedrow.

The Julliere model [3]
In 1975 Julliere had an idea to replace the superconductor by another ferromagnet and thus obtain a magnetic tunnel junction (MTJ) of the structure F/I/F. Instead of using a S as a spin detector he used the exchange-split states of another F. The tunneling current then depends on the relative magnetization orientation of the two F electrodes: the parallel (P) state has low resistance, \( R_P \), and the anti parallel (AP) state has high resistance, \( R_{AP} \). The majority (minority) electrons from the first F tunnel into the available majority (minority) states in the second F which in the P state are equal in number. In the AP state, on the other hand, the populations are reversed and the available states in the second F become fewer. As a result, the electrons encounter higher resistance. This is in analogy to light polarization where two polarizing plates will either block or transmit light, depending on the relative orientation of the plates. The normalized difference in the resistances is defined as the tunneling magneto resistance (TMR):

\[
TMR = \frac{R_{AP} - R_P}{R_P}.
\]

Using Co and Fe and a tunnel barrier, Julliere observed a TMR of about 14% at 4 K.

Julliere assumed spin conserved tunneling and that the number of conduction electrons in electrodes \( F_1 \) and \( F_2 \) is proportional to the density of states at the Fermi energy, \( N^{1(2)}(E_F) \). The spin-up and spin-down conduction electrons in the electrodes have

\[

case{N^{1(2)}_\uparrow}{a^{1(2)} N^{1(2)}_\uparrow},
case{N^{1(2)}_\downarrow}{(1 - a^{1(2)}) N^{1(2)}_\downarrow},
\]

where \( a^i \) is the fraction of the spin-up conduction electrons in electrode \( i = 1, 2 \). Accordingly, the conductance for the P and AP alignments are

\[
\begin{align*}
G_P &\propto (N^{1}_\uparrow N^{2}_\uparrow + N^{1}_\downarrow N^{2}_\downarrow), \\
G_{AP} &\propto (N^{1}_\uparrow N^{2}_\downarrow + N^{1}_\downarrow N^{2}_\uparrow).
\end{align*}
\]

It follows from Eq. 3.11 and 3.12 that the conductances are different for the different magnetic states, which gives a non-zero TMR. Eq. 3.8 results in the following
expression for the TMR

\[ TMR = \frac{G_P - G_{AP}}{G_{AP}} = \frac{2(2a_1 - 1)(2a_2 - 1)}{1 - (2a_1 - 1)(2a_2 - 1)} = \frac{2P_1 P_2}{1 - P_1 P_2}, \] (3.13)

where \( P_1 \equiv 2a_1 - 1 \) and \( P_2 \equiv 2a_2 - 1 \). The disadvantage of this model is that it does not take into account the height nor the thickness of the barrier but only the DOS at \( E_F \). As a consequence, the voltage bias dependence of the TMR is not explained by this model.

The Slonczewski model [53]

A more accurate theoretical consideration of TMR was presented by Slonczewski in 1989 where he included the height and thickness of the barrier. The model was based on the one-electron Hamiltonian within the free electron approximation and on Stearns [52] theory of s-d hybridized bands. Assuming a rectangular barrier with matching spin dependent electron wave functions across the junction, Slonczewski solved the Schrödinger equation for a F/I/F junction. As a result, the conductance as a function of the relative magnetization alignment (\( \theta \)) of the two F’s was obtained to be

\[ G(\theta) = G_0 \left(1 + P^2 \cos \theta\right), \] (3.14)

where \( G_0 \) is the mean surface conductance, and the \( P \) is the effective spin polarization of the tunneling electrons

\[ P = \left(\frac{k_\uparrow - k_\downarrow}{k_\uparrow + k_\downarrow}\right) \cdot \left(\frac{\kappa^2 - k_\uparrow k_\downarrow}{\kappa^2 + k_\uparrow k_\downarrow}\right). \] (3.15)

Here \( \kappa \) is a constant that depends on the barrier height \( V \), \( \kappa = \sqrt{\left(\frac{2m}{\hbar^2}\right)(V - E_F)} \). Observe that, in the case of a high barrier, Slonczewski’s result for the TMR is reduced to that of Jullier’s.

The fact that the polarization depends on the barrier height is a direct indication that the SP of the conduction electrons is not an intrinsic property of the F’s band structure. On the other hand, the model does not describe voltage, temperature or thickness dependence of the MR. In fact, MacLaren et al. [54] compared the Julliere model [3] and Slonczewski’s model [53] with the exact expression for the magneto conductance of free electrons and a numerical calculation for band electrons in Fe based tunnel junctions. They concluded that neither model is appropriate for describing the tunneling between two Fe films. However, Slonczewski’s model does provide a good approximation in the thick barrier limit and for small barrier heights. A realistic and accurate model should incorporate true electron band structure including the interfaces. Moreover, it should take into account the influence of the barrier height and thickness on the tunneling current.
3.4 Spin Polarized Transport

We have now discussed the possibility to create a spin polarized current with the help of ferromagnetic electrodes. The next task is to investigate the possibility to transport this current in a non-magnetic material. Already in 1963 Clark and Feher [55] demonstrated a method for generating non-equilibrium spin in InSb. The experiment was based on the Feher effect where they drove a current through the sample while applying a constant magnetic field. A decade later Aronov [56, 57] and Pikus [8] proposed a method for spin injection from a F into metals, semiconductors and superconductors. They predicted that such a spin polarized current injection would lead to a non-equilibrium magnetization in the non-magnetic material at the F/N interface. It was not until a decade later electrical spin injection and precession into a metal was first realized by Johnson and Silsbee [9–11] in their pioneering experiments on single crystal aluminum samples. Figure 3.3 shows the non-local measurement setup of the experiment, where the current is sent towards one side of the sample through a Py pad and voltage is measured on the other side, with another Py pad. This way, they measured a weak (≈ 70 pV) spin induced signal using a SQUID at temperatures up to 77 K.

Spin injection in F/N junctions has been subsequently studied in detail by Johnson and Silsbee [10, 58], van Son et al. [59], Valet and Fert [60], Hershfield and Zhao [61], Rashba [62,63], Jedema et. al [12,13], Takahashi and Maekawa [64], Garzon et. al. [14], Valenzuela et. al [15, 16], Urech et. al [17], Appelbaum et. al. [18] and others. Johnson and Silsbee based their model on thermodynamic considerations whereas Valet and Fert applied the Boltzmann transport equation to obtain equivalent results. Below, we follow the discussion by Rashba.

Recall that the DOS in a normal metal are not shifted, i.e the spin-up and spin-
Figure 3.4: Schematic representation of the DOS of a ferromagnet (left) and a normal metal (right). Injection of spin polarized electrons from the ferromagnet will cause a spin accumulation in the normal metal in the vicinity of the interface.

down conductivities are equal. As a consequence, injecting spin polarized electrons into N will result in a spin accumulation over a certain distance, on either side of the F/N interface. This will create an unbalance in the DOS of the N, illustrated in Figure 3.4, which is associated with different electro chemical potentials, $\mu_\uparrow$ and $\mu_\downarrow$. By definition, the magnitude of this difference ($\mu_\uparrow - \mu_\downarrow$) is the spin accumulation.

3.5 Spin Injection and Diffusion

Consider a one dimensional system consisting of an infinite long non-magnetic metal wire with an F contact at $x = 0$. The spin current density in N is proportional to the space derivative of $\Delta \mu$:

$$j_s = -\frac{\sigma_N}{e} \nabla (\Delta \mu),$$  \hspace{1cm} (3.16)

where $\sigma_N$ is the conductivity in N and $\Delta \mu = (\mu_\uparrow - \mu_\downarrow)/2$ is the splitting in the electro chemical potentials. Moreover, $\Delta \mu$ is proportional to the difference in spin-up and spin-down electron densities $n_\uparrow$ and $n_\downarrow$ according to

$$\Delta \mu = \frac{D}{\sigma_N} (n_\uparrow - n_\downarrow) \equiv \frac{D}{\sigma_N} n_s,$$  \hspace{1cm} (3.17)
3.5. SPIN INJECTION AND DIFFUSION

where $D$ is the diffusion constant in $N$. In the steady state, particle conservation leads to the continuity equations

$$\frac{\partial n_1}{\partial t} = 0 \Rightarrow \nabla j_1 = \frac{n_1}{\tau_{\uparrow\uparrow}} + \frac{n_1}{\tau_{\downarrow\downarrow}},$$

$$\frac{\partial n_1}{\partial t} = 0 \Rightarrow \nabla j_1 = + \frac{n_1}{\tau_{\uparrow\downarrow}} - \frac{n_1}{\tau_{\downarrow\uparrow}},$$

(3.18)

where $\tau_{\sigma\sigma'}$ is the average time for the spin to flip from $\sigma$ to $\sigma'$. In $N$, the two spin relaxation times are equal, i.e $\tau_{\uparrow\downarrow} = \tau_{\downarrow\uparrow} = \tau_{sf}$. Combining Eqs. 3.16, 3.17 and 3.18 and using the Einstein relation (Eq. 3.1) the spin diffusion processes can be described by the following diffusion equation:

$$\frac{\partial^2}{\partial x^2} \Delta \mu = \frac{1}{\lambda_{sf}} \Delta \mu,$$

(3.19)

where $\lambda_{sf} = \sqrt{D \tau_{sf}}$ is the spin diffusion length. Note that this equation is independent of the charge current and therefore describes the diffusion of spins only. Figure 3.5 (a) illustrates the fact that the spin diffusion in $N$ is isotropic irrespective of the charge current that only flows in $x < 0$. In order to maintain charge neutrality, when a spin-up current flows in the positive direction, a spin-down current of equal magnitude flows in the negative direction, see Figure 3.5 (b).

Figure 3.5: (a) Crossover of the 1-D geometry where two ferromagnets act as spin injector (F1) and spin detector (F2). Only spin current flows in the detector path ($x > 0$). (b) Spatial variation of the electrochemical potential and spin current for spin-up and spin-down electrons in $N$. After [65].
CHAPTER 3. SPIN DEPENDENT TRANSPORT

3.6 Spin Accumulation

For a 1-D infinitely long wire with thickness $t$ and width $w$ ($A = wt$) the solution of the diffusion equation is

$$\Delta \mu(x) = \Delta \mu_0 \exp(-|x|/\lambda_{sf}),$$  \hspace{1cm} (3.20)

where $\Delta \mu_0$ is the spin splitting at the interface and is determined by combining Eqs. 3.16 and 3.19:

$$\Delta \mu_0 = e \gamma j \lambda_{sf} / \sigma N I.$$  \hspace{1cm} (3.21)

Since the spins are diffusing in both directions, $x < 0$ and $x > 0$, the spin splitting at the injection point is halved, $\Delta \mu_0 \rightarrow \Delta \mu_0 / 2$, i.e.

$$\Delta \mu(x) = \frac{\gamma \lambda_{sf}}{2 e \sigma N A} I \exp(-|x|/\lambda_{sf}).$$  \hspace{1cm} (3.22)

The polarization of the injected current is $[58, 62, 63]$

$$\gamma = \frac{P r_c + p_F r_F}{r_c + r_N + r_F},$$  \hspace{1cm} (3.23)

where $r_c = [G(1 - P^2)]^{-1}$ is the effective interface resistance, $P$ the interface polarization, $r_F = \lambda_F / \sigma_F (1 - p_F^2)$ the characteristic resistance over one spin diffusion length ($\lambda_F$) in $F$, and $r_N = \lambda_{sf} / \sigma_N$ the characteristic resistance in $N$.

By examining Eq. 3.23 one can investigate the possibilities for effective spin injection into $N$. For example, using an ideal metallic contact ($r_C = 0$) with a resistance mismatch due to the difference in spin diffusion lengths in the materials, $r_F << r_N$, leads to smaller injection efficiency, with $\gamma \approx r_F / r_N << 1$. Accumulated spins at the interface will diffuse even into $F$ and due to the short spin diffusion length in $F$ ($\lambda_F << \lambda_N$) their spin information will be lost quickly. The ferromagnet will act as a spin sink for the back flowing electrons.

Employing resistive contacts should lead to more efficient spin injection. The perfect candidate is a tunnel junction which generally has much larger resistance than $r_N$ and $r_F$. The barrier will prevent tunneling electrons from scattering back into $F$ and loosing their spin orientation. The electrodes will only act as injecting and detecting probes and their proximity will not disturb the spin accumulation in $N$. The current polarizations then simply becomes equal to the interface polarization, i.e $\gamma = P$.

It is not crucial for the junction resistance, $r_C$, to be very large. In fact, the necessary criterium is that it should be larger than the effective resistances, e.g $r_C >> \lambda_F / \sigma_F$. The original treatment of this resistance mismatch problem belongs to Johnson and Silsbee [58, 66], who work out a general expression for the injection efficiency, taking into account the interface resistance and spin asymmetry. This interface or contact resistance is typically non-zero even for metallic contacts, and can be sufficient to fulfill the above criterion for efficient spin injection. As
3.7. MECHANISMS OF SPIN RELAXATION

Godfrey and Johnson demonstrate in a recent publication [67, 68], the contact resistance even for metallic contacts with specially prepared clean F/N interfaces can be comparable with the effective normal and ferromagnetic resistances, which determine the injection efficiency in the spirit of Eq. 3.23. This contact resistance then minimizes the unwanted effect of spin-sinking into the ferromagnetic injector. Other experiments by Otani et. al. [69] and Poli et al. (in chapter spin absorption by ferromagnets) report significant spin sinking effects for ferromagnetic metallic junctions. The results will likely vary for different materials and fabrication methods. The consensus appears to be that a certain optimum interface resistance is desirable, and its range is determined by the transport parameters of the specific device.

3.7 Mechanisms of Spin Relaxation

We have already mentioned that the spin accumulated at the interface will relax according to the diffusion Eq. 3.19, but the responsible mechanisms for this effect are yet to be discussed. In order to alter a magnetic moment such as spin, some kind of magnetic influence is needed. The Bloch-Torrey equations [70,71] can be used to describe the dynamics of mobile electrons, their spin relaxation and spin dephasing. In the presence of an applied magnetic field \( \mathbf{B}(t) = B_0 \hat{z} + B_1(t) \), the spin relaxation time \( T_1(\tau_{sf}) \) and spin dephasing time \( T_2 \) are defined via the equations

\[
\frac{\partial \mathbf{M}}{\partial t} = \gamma (\mathbf{M} \times \mathbf{B}) + D \nabla^2 \mathbf{M} - \left[ \begin{array}{c} M_x/T_2 \\ M_y/T_2 \\ (M_z - M_0)/T_1 \end{array} \right],
\]

where \( \gamma = \mu_B g / \hbar \) is the electron gyro magnetic ratio, \( D \) is the diffusion constant and \( M_0 = \chi B_0 \) the thermal equilibrium magnetization, with \( \chi \) being the systems susceptibility.

In the absence of magnetic impurities, the dominant spin relaxation mechanism in metals is due to the spin-orbit interaction which was derived independently by Elliot [72] and Yafet [73]. They realized that the spin of conduction electrons can relax via ordinary momentum scattering in the presence of spin-orbit coupling induced by the lattice ions. This is a relativistic effect and is explained by considering the moving electron in its rest frame, where the periodic lattice potential is transformed into a magnetic field, i.e

\[
V_{SO} = \frac{\hbar}{4m^2c^2} (\nabla \mathbf{U} \times \hat{p} \cdot \hat{\sigma}). \tag{3.25}
\]

Here, \( m \) is the free electron mass, \( \mathbf{U} \) is the periodic lattice potential, \( \hat{p} \equiv -i\hbar \nabla \) is the linear momentum operator and \( \hat{\sigma} \) are the Pauli matrices. The electron Bloch eigenstates become linear combinations of the spin-up \(|\uparrow\rangle\) and spin-down \(|\downarrow\rangle\) states.
ψ_{kn\uparrow}(\mathbf{r}) = [a_{kn}(\mathbf{r}) |\uparrow\rangle + b_{kn}(\mathbf{r}) |\downarrow\rangle] e^{i k \cdot \mathbf{r}}, \quad \text{(3.26)}

ψ_{kn\downarrow}(\mathbf{r}) = [a^*_{-kn}(\mathbf{r}) |\downarrow\rangle - b^*_{-kn}(\mathbf{r}) |\uparrow\rangle] e^{i k \cdot \mathbf{r}}, \quad \text{(3.27)}

where $k$ is the lattice momentum, $n$ is the band index and $a_{kn}(\mathbf{r})$ and $b_{kn}(\mathbf{r})$ are the complex periodic coefficients that depend on the radius vector $\mathbf{r}$. Often, the spin-orbit interaction can be incorporated into the band structure as a perturbation giving $|b| \approx \lambda_{SO}/\Delta E$, where $\lambda_{SO}$ is some effective spin-orbit parameter and $\Delta E$ is the energy difference between neighboring bands. Typically, the spin-orbit interaction is weak and much smaller than $\Delta E$ leading to $|b| \ll 1$. Its strength depends on the atomic number ($\propto Z^4$) which for example explains why $\lambda_{sf}$ in Au is much shorter than in Al (Au is much heavier).

In general, the spin-orbit interaction alone does not lead to spin relaxation. However, in combination with momentum scattering it is possible for the electron to flip its spin. The polarized electrons scatter around approximately $10^4$ times before they are relaxed in spin. In other words, the spin relaxation time is much longer than the momentum relaxation time. The momentum scattering is caused by several factors as discussed in section Thin Film Resistivity, such as magnetic and non-magnetic impurities, surfaces and grain boundaries. This means that the resistivity $\rho$ of the material is also a very important parameter as can be seen from the simple relation derived by Yafet [73]:

$$\frac{1}{\tau_{sf}} \sim b^2 \rho. \quad \text{(3.28)}$$

At high temperatures (above the Debye temperature $T_D$) where phonon scattering dominates we have $1/\tau_{sf} \sim \rho \sim T$. At lower temperatures, the phonon scattering becomes negligible and Yafet expects the spin relaxation to depend on $T$ according to $1/\tau_{sf} \sim T^5$ which is similar to the Bloch-Gruneisen law for resistivity. This is difficult to observe since in most cases the scattering is dominated by impurities, grains and surfaces.

Fabian and Das Sarma [74–76] solved the question why very similar metals may have spin relaxation rates differing by 2 or 3 orders of magnitude by performing ab initio pseudo potential band structure calculation on Al. They found that spin relaxation in metals is significantly enhanced whenever a Fermi surface crosses Brillouin boundaries, special symmetry points or lines of accidental degeneracy. Although these regions, even called spin hot spots, comprise only a small part of the Fermi surface they have enough weight to dominate the spin relaxation. This explains why the spin relaxation rates for Al and Na differ by 2 orders of magnitude although atomically they differ only by 10%. The reason is that the Fermi surface of Na is nearly spherical and lies entirely within the first Brillouin zone. On the other hand, the Fermi surface of polyvalent Al is more complex and crosses Brillouin zone boundaries, leading to spin hot spots and a higher spin relaxation rate.
3.8 Spin Detection

The non-equilibrium spin discussed in the previous section decays exponentially along the wire. Placing another ferromagnet (F2) at position \(x\) along the wire enables us to detect \(\Delta \mu\) at that distance. The voltage drop \(V\) across the detector junction is due to extremely small spin currents that tunnel in and out of F in order to maintain charge neutrality. Hence only a pure spin current flows through the detector junction where the measured potential is a weighted average of \(\mu^\uparrow\) and \(\mu^\downarrow\). Depending on the polarization parameter of the detector, \(P\), which is assumed to be equal to that of the injector, the voltage is given by [11]

\[
eV_{P(AP)} = \pm P \frac{n_s}{N(E_F)} = \pm P \Delta \mu. \tag{3.29}
\]

Here \(+\)\((-\) stands for the parallel (anti-parallel) magnetization configuration of the F electrodes. The total spin valve signal which is the voltage difference between the parallel and anti-parallel state is given by \(V_S = V_P - V_{AP} = 2P \Delta \mu/e\).

Combining this result with Eq. 3.19 results in the spin signal [9,59–64]

\[
R_S = V_S/I = P^2 R_N e^{-|x|/\lambda_{sf}}, \tag{3.30}
\]

where \(R_N = \lambda_{sf}/\sigma_N A\).

When both junctions are transparent contacts we have [12,61–63]

\[
R_S = \frac{4P_F^2}{(1 - P_F^2)^2} R_N \left( \frac{R_F}{R_N} \right)^2 \frac{e^{-|x|/\lambda_{sf}}}{1 - e^{-2|x|/\lambda_{sf}}}. \tag{3.31}
\]

When one of the junctions is a transparent contact and the other is a tunnel junction we have

\[
R_S = \frac{2P_F P}{(1 - P_F^2)} R_N \left( \frac{R_F}{R_N} \right) e^{-x/\lambda_{sf}}. \tag{3.32}
\]

Here \(R_F = \rho_F \lambda_F/A_F\), \(A_F\) is the area of the junction. The resistance mismatch factor \((R_F/R_N)\) is removed when a transparent contact is replaced with a tunnel junction. Thus the maximum spin signal is achieved when all contacts are made of tunnel junctions.

**Symmetric spin diffusion**

Charge and spin are intrinsic and fundamental properties of individual electrons, and can not be separated by any means. However, for an ensemble of electrons, this spin-charge coupling can be broken. It follows from the fact that the charge is strictly conserved, whereas the spin is not, due to spin-flip scattering. An example of such an ensemble is the spin \((J_S)\) and charge \((J_C)\) currents, created by current injection from a ferromagnet into a non-magnetic metal. In fact, a device like ours, is a perfect candidate for demonstrating that these two currents can be decoupled.
Figure 3.6: Schematic illustration of isotropic spin diffusion. The charge flows only to the right side in the wire, whereas the spin current flows both ways. The spin accumulation is measured by two identical detectors ($D_1$ and $D_2$), on either side of the injector.

The width and thickness of the Al strip is much smaller than $\lambda_{sf}$, making it a 1D channel for spin diffusion.

Figure 3.6 shows the schematic illustration of the spin and charge transfer for a current injected in a non-magnetic nano-wire. The density of the charge current is uniform and flows from the injector ($I$) and to the right side in the wire. On the other hand, the density of the spin current decays exponentially and flows symmetrically about the injection point. Two identical detectors, at equal distances and on either side of the injector, are then used to detect the non-equilibrium spin accumulation. Observe that one of the detectors is placed within the current path ($D_2$) whereas the other one ($D_1$) is placed outside the current path. Nevertheless, the spin accumulated at either detector should be equal, irrespective of the presence of the charge current. This device allows us to directly demonstrate that spin and charge currents are strictly decoupled in diffusive transport channels.
Chapter 4

Spin Injection, Accumulation and Relaxation in Superconductors

4.1 Superconductivity

Spin injection into superconductors (S) is more delicate than for normal metals (N). The physical properties of a S can be easily perturbed through different mechanisms, such as temperature, current, and magnetic field. It is therefore important to setup an experiment that avoids the un-deliberate effects on S. In this chapter, I will introduce the relevant properties of a S and discuss our experiments using F/S nano structures.

4.2 Introduction

Superconductivity is one of the most interesting phenomena in physics. It was discovered by the Dutch physicist Heike Kamerlingh Onnes in 1911, when he cooled mercury to liquid helium temperature (4 K) and found that its resistance dropped to zero at 4.1 K. He obtained infinite conductivity, i.e. the current flowed in the superconductor without any resistance. A simple physical explanation for this effect is as follows: as an electron passes by the positive ions in the lattice, it will attract the ions towards it. This deformation of the lattice results in a higher density of positive ions at that point. A second electron will be attracted towards the higher density of positive ions, thus experiencing an effective attractive interaction with the first electron. If this attraction overcomes the Coulomb repulsion, a net attractive interaction is created, and the two electrons are bound together into a Cooper pair. Such stable electron pairs are formed only of electrons of opposite spins, since the electrons are fermions and obey the Pauli exclusion principle. Hence, Cooper pairs have zero net spin and fall in the category of bosons. This makes it possible for the Cooper pairs to condense into a common ground state, i.e. Bose-Einstein condensate, that posses infinite electrical conductivity.
Cooper pairs cannot be used for spin transport in S, since they carry no spin. However, the unpaired electrons in the excited states of the superconductor, referred to as quasi-particles (QP), have fermionic nature. They carry spin $1/2$, which makes them suitable as spin carriers. In our experiments, we study spin transport by such QP’s in F/S nanostructures.

Another characteristic property of a S is perfect diamagnetism, also called the Meissner effect. External magnetic field lines are expelled from S by super-currents flowing in a very thin outer shell of the superconducting body. These currents produce a magnetic field which counters the external field. If the applied magnetic field is stronger than a certain critical value, $H_c$, the superconductivity is destroyed through magnetic field induced depairing of the Cooper pairs. High current densities carried by a superconductor can also drive the S normal state. Therefore, the electromagnetic environment in F/S experiments must be considered carefully.

4.3 The Bogoliubov-de Gennes Equation

A quasi-particle in S is an un-paired electron which can have one of two characters, electron like $|e\rangle$ or hole like $|h\rangle$, depending on the excitation energy. It can be described by the wave function

$$\Psi(x, t) = a(x, t) |e\rangle + b(x, t) |h\rangle,$$

where $a$ and $b$ are the probability amplitudes to find the QP in $|e\rangle$ or $|h\rangle$, respectively. The wave function obeys the Bogoliubov-de Gennes equation

$$i\hbar \frac{\partial \Psi}{\partial t} = \left[ \begin{array}{cc} H & \Delta \\ \Delta & -H \end{array} \right] \Psi,$$

where

$$H = -\frac{\hbar^2}{2m} \frac{d^2}{dx^2} - E_F + V(x),$$

$E_F$ is the Fermi energy and $V$ is the interaction potential between the QP and the lattice. The interpretation of Eq. 4.2 is that the QP in the state $|e\rangle$ obeys the Schrödinger equation whereas the QP in the state $|h\rangle$ obeys the time reversed Schrödinger equation. Separating the wave function in time and space, i.e $\psi(x, t) = \psi(x)e^{-i\omega t}$, and using the trial solution

$$\psi = \left[ \begin{array}{c} a \\ b \end{array} \right] e^{ikx},$$

one finds that

$$E_k = \sqrt{\epsilon_k^2 + \Delta^2}.$$
where \( \epsilon_k = \left( \frac{\hbar^2 k^2}{2m} - E_F \right) \) is the energy relative to \( E_F \) of an electron with momentum \( k \) in the normal state. If \( |k| \) is larger than the Fermi wave vector \( k_F \) the QP has the electron like character, i.e positive root of Eq. 4.5. In the opposite case it has the hole like character, i.e negative root of Eq. 4.5. The two bands are essentially linear, except for when \( k \approx k_F (E \approx \Delta) \), where an energy gap of \( 2\Delta \) is present. Hence, depending on the excitation energy of the QP’s they will be either in the \( |e\rangle \) or \( |h\rangle \)-band respectively. This can create a difference in the population of the bands which is often referred to as charge imbalance and will be discussed ahead. At equilibrium, the bands are equally filled and charge neutrality is maintained.

4.4 BCS Superconductivity

A well known study of superconductivity has been performed by Bardeen, Cooper and Schrieffer [77]. In their formulation, the ground state is formed by Cooper pairs \(|k, \uparrow; -k, \downarrow\rangle\) consisting of two electrons opposite momentum and spin. The probability of the ground state being empty or occupied is \(|v_k|^2\) or \(|u_k|^2\), respectively, with \(|v_k|^2 + |u_k|^2 = 1\). The Cooper pairs carry no momentum in the center of mass nor any spin, but only charge (2e). On the other hand the QP’s carry spin and have non-integer charge. They can be expressed as a linear combination of the creation and annihilation operators of normal electrons, \(c_{k,\sigma}^+\) and \(c_{k,\sigma}\) [78],

\[
\begin{align*}
\gamma_{k,\uparrow} &= u_k c_{k,\uparrow}^+ - v_k c_{-k,\downarrow}^+ \\
\gamma_{-k,\downarrow} &= u_k c_{-k,\downarrow}^+ + v_k c_{k,\uparrow}^+
\end{align*}
\]

The operators \(\gamma_{k,\sigma}\) create QP excitations of the two spin directions from the superconducting ground state. Creating a hole like QP is equivalent to annihilating a pair and creating an electron like QP. Moreover, the fractional effective charge of the QP’s is

\[
q_k = u_k^2 - v_k^2 = \frac{\epsilon_k}{E_k},
\]

where the BCS coherence factors are given by

\[
\begin{align*}
|u_k|^2 &= \frac{1}{2} (1 + \frac{\epsilon_k}{E_k}) \\
|v_k|^2 &= \frac{1}{2} (1 - \frac{\epsilon_k}{E_k})
\end{align*}
\]

The QP excitations change in character from the electron like (with effective charge \( q_k \approx 1 \)) to hole like (\( q_k \approx -1 \)) as the excitation moves in energy from outside the Fermi surface to the inside.
CHAPTER 4. SPIN INJECTION, ACCUMULATION AND RELAXATION IN SUPERCONDUCTORS

As mentioned previously, the basic property of a superconductor is the energy gap $\Delta$ in the DOS, $N_S(E)$, within which there is no available states:

$$D_S(E) = \frac{N_S(E)}{N_N} = \begin{cases} \frac{E}{\sqrt{E^2 - \Delta^2}} & (E > \Delta) \\ 0 & (E < \Delta) \end{cases},$$  \hspace{1cm} (4.11)$$

where $N_N$ is the DOS in the normal state. Figure 4.1 shows the DOS of the QP’s at $T = 0$, compared to the normal state. Observe that there are no available states for QP’s with energies less than $\Delta$. Instead, these states are distributed non uniformly for $E \geq \Delta$. Different superconductors have different $\Delta$, and for Al it is approximately $200 \ \mu$ eV. The temperature dependence of the energy gap is given by the self consistency condition [78]

$$\frac{1}{V} = \frac{1}{2} \sum_k \frac{\tanh(\beta E_k/2)}{E_k},$$  \hspace{1cm} (4.12)$$

where $\beta = 1/kT$, $E_k$ is given by Eq. 4.5 and $V$ is some scattering potential. At very low temperatures, $\Delta$ is nearly constant until a significant number of QP’s are thermally excited which makes it drop to zero near $T_C$, approximately as $\Delta(T)/\Delta(0) \approx 1.74 \sqrt{(1 - T/T_C)}$. It is known that injection of high energy QP’s suppress the gap [79], which will be found also in our results.
4.5 Normal Metal-Superconductor (N/I/S) Tunneling

Before discussing tunneling in N/I/S junctions, we mention the main characteristics of metallic N/S contacts. In this case, transport is possible via the Andreev reflection process [80]. Since there are no available states for un-paired electrons below $\Delta$, incident electrons with $E < \Delta$ are transmitted into S as Cooper pairs, while a hole is reflected back into N. Each electron transfers twice the charge, doubling the conductance compared to the normal state. This was considered by Blonder, Tinkham and Klapwijk [81] who studied the transport properties in a N/I/S junction as a function of barrier strength ranging from a metallic contact to a low transparency barrier. For a metallic contact, they found that the sub-gap conductance was twice the normal state conductance. Another effect is the proximity effect in which Cooper pairs from S diffuse into N. Thirdly, in the case of F/S interface, suppression of superconductivity is possible due to the ferromagnetic exchange interaction at the interface. These unwanted effects in metallic contacts are negligible for a tunnel barriers. Therefore, we use tunnel junctions in our experiments.

Electron tunneling provides an excellent experimental examination of the DOS of a metal. This technique was pioneered by Giaever [82] who demonstrated that the tunnel conductance of a N/I/S junction directly probes the DOS of a superconductor. The differential conductance reflects the shape of the single electron DOS of the superconductor. When a voltage is applied across a N/I/S junction, electrons will start to tunnel whenever the magnitude is larger than the gap energy. This is illustrated in Fig. 4.2 where the DOS of the N and S are represented in the semiconductor model and the corresponding current-voltage characteristic is shown. For $eV < \Delta$, electrons can not tunnel into S, since there are no available states in S, which results in a zero current through the junction. On the other hand, if the voltage is raised such that $eV \approx \Delta$ the electrons will be able to tunnel into the free QP states in S and one observes a sudden increase in current. At negative bias, the energy in S is raised with respect to that in N until it reaches $E_F$ at $-\Delta$. The free states in N are now available for broken Cooper pairs, which allows tunneling to occur. As a consequence, the I-V curve is symmetric and clearly demonstrates the energy gap. For higher voltages, the I-V curve approaches the normal state behavior since the influence of the gap on the tunnel current becomes negligible at $E >> \Delta$. The dashed line in Figure 4.2 (b) represents $T = 0$ with a pronounced step, corresponding to the superconducting gap.

The net tunneling current into the superconductor is given by [78]

$$I_{N\rightarrow S} = \frac{G_{NN}}{e} \int_{-\infty}^{\infty} D_S(E) [f(E) - f(E + eV)] dE,$$  \hspace{1cm} (4.13)

where $G_{NN}$ is the tunnel conductance of the junction when both metals are in the N state, $D_S(E)$ is the normalized BCS DOS (Eq.4.11) and $f(E)$ is the Fermi
CHAPTER 4. SPIN INJECTION, ACCUMULATION AND RELAXATION IN SUPERCONDUCTORS

Figure 4.2: (a) Tunneling between a normal metal and a superconductor (N/I/S) represented in the semiconductor model. (b) Current-Voltage characteristic curve of a N/I/S junction.

distribution function

\[ f(E) = \frac{1}{\exp(E/kT) + 1} \tag{4.14} \]

For a more direct comparison between experiment and theory, the differential conductance, \( dI/dV \), is normally measured. At low temperatures, it is directly proportional to the superconducting DOS, \( D_S(E) \). The ratio of the conductance at \( T < T_C \) to the conductance in the N-state is,

\[ \frac{G_{NS}}{G_{NN}} = \int_{-\infty}^{\infty} D_S(E) \left[ \frac{\kappa e^{\kappa(E+eV)}}{(1+e^{\kappa(E+eV)})^2} \right] dE, \tag{4.15} \]

where \( \kappa = 1/kT \).

The injected QP’s will relax in energy to the gap edge, giving up their excess energy \( (\approx 2\Delta) \) through different mechanisms, before recombining into the condensate. The recombination time \( \tau_R \) is very long, typically of the order of \( \mu s \) in Al [83, 84]. On a shorter time scale \( (\tau_E) \) occurs the QP energy relaxation which in general is due to electron-phonon, electron-electron and electron-impurity scattering. For most materials, the dominant interaction is due to electron-phonon processes, except for metals with high Debye frequency such as Al and Zn [85,86], for which electron-electron scattering processes can be significant. In fact, the energy relaxation time due to the electron-phonon processes can be \( \tau_{e-ph} \approx 5 \cdot 10^{-7} \) s [85], whereas for electron-electron processes it is \( \tau_{e-e} \approx 4 \cdot 10^{-7} \) s. Since both time scales are much longer than the spin relaxation time \( (\approx 100 \text{ ps}) \), relevant spin-dependent transport is often modeled in the elastic regime [87], which is the
4.6. CHARGE IMBALANCE

As mentioned earlier, the injected electrons have the probability $u_k^2$ of entering the $|e\rangle$-branch and $v_k^2$ of entering the $|h\rangle$-branch. In thermal equilibrium, these two probabilities are equal and the chemical potential of the Cooper pairs $\mu_{\text{CP}}$ is equal to the chemical potential of the QP’s, $\mu_{\text{QP}}$. The excitation spectrum of the QP’s is given by Eq. 4.5 and can be shifted towards either branch depending on the magnitude and sign of the applied voltage. If $eV \gg (\Delta, k_B T)/e$ then $u_k^2 \gg v_k^2$ and the majority of the injected QP’s will be in the $|e\rangle$-branch, see Figure 4.3. The situation is reversed for negative voltage and the $|h\rangle$-branch becomes preferentially populated. For small voltages, $eV \sim \Delta$, $u_k^2 \sim v_k^2$ and the branches are equally populated.

It is the difference between these two branches that is referred to as branch imbalance or charge imbalance and has been extensively investigated by a number of groups [86, 88–94]. The charge imbalance per unit volume is given by [89]

$$Q = n_> - n_< = 2N(0) \int_{\Delta}^{\infty} D_S(E) (f_{k>} - f_{k<}) , \tag{4.16}$$

where $N(0)$ is the density of states for electrons of one spin, $D_S(E)$ is the normalized BCS DOS (Eq.4.11) and $f_{k>}$ and $f_{k<}$ are the steady state non-equilibrium population of the $|e\rangle$- and $|h\rangle$-branch, respectively. In order to maintain overall electrical neutrality, there must be a compensating change in the number of
CHAPTER 4. SPIN INJECTION, ACCUMULATION AND RELAXATION IN SUPERCONDUCTORS

Cooper pairs, i.e. the electro-chemical potentials \( \mu_{QP} \) and \( \mu_{CP} \) will shift in opposite directions. This results in a measurable potential difference which has been demonstrated by Tinkham and Clarke [88, 89] to be given by

\[
V = \frac{1}{e g_{NS}} \int_{-\infty}^{\infty} (f_{k>} - f_{k<}),
\]

(4.17)

where \( g_{NS} = G_{NS}/N_N \) is the normalized tunneling conductance of the junction.

The charge imbalance will relax in space exponentially as \( \sim e^{-x/\lambda_Q} \), where \( \lambda_Q = \sqrt{D \tau_Q} \) is the appropriate diffusion length during time \( \tau_Q \). The relaxation can be due to inelastic scattering, annihilation and creation processes through phonons. Gap anisotropy and elastic scattering can also relax \( Q \). To summarize, charge imbalance can be neglected at small injection energies. This is the regime where we will focus our attention experimentally.

4.7 Ferromagnet-Superconductor (F/I/S) Tunneling

So far, we have only treated non-polarized spin injection into S using N/I/S junctions. The situation becomes much more complex and interesting if N is replaced by F, in which case we are back to spin polarized current injection. Once again, Johnson [95] was the first to perform an experimental study on spin injection and detection in S (Nb), where he used the non-local measurement technique discussed earlier. He found a strong reduction of \( \lambda_{sf} \) at \( T < T_C \). Later, Gu et al. [96] inferred a much smaller reduction using local measurements across a metal stack containing Nb. Recently, similar experiments have been made [97-99] where in reference [98], the authors injected spin with only one F electrode and measured the resistive fraction in S using several N electrodes. This way they inferred a two-fold decrease of \( \lambda_{sf} \) in Al below \( T_C \). However, all these experiments on spin injection in S used metallic contacts between the F electrodes and S, which is known to lead to proximity effects and/or strong Andreev processes [80]. We avoid this by using tunnel contacts. Furthermore, \( \lambda_{sf} \) was not measured directly in previous experiments as the magnitude of spin splitting in S versus the distance from the spin injection point. Our multi-electrode device enables us to detect the spin signal simultaneously at different distances from the injection point. This allows a direct measurement of the spin relaxation parameters \( (R_S, \lambda_{sf}) \) in S.

Several theories of the tunneling and relaxation of spin-polarized QP’s in S have been developed recently [64, 100-104]. We first follow the approach by Takahashi et. al. [64,101, 102] to obtain an expression for the spin-polarized QP current in S, in the non-local geometry, illustrated in Figure 6.5. Neglecting contribution from charge imbalance (small \( eV \)) and considering the in-elastic regime, i.e \( \tau_E < \tau_{sf} \), they calculate the spin-dependent tunnel currents across a F/I/S junction to be

\[
I_{\uparrow(\downarrow)}(V) = \frac{G_{T \uparrow(T \downarrow)}}{e} \left[ \int_{-\infty}^{\infty} D_S(E) [f(E - eV) - f(E)] dE \right] - (+)S,
\]

(4.18)
4.8 Spin Accumulation and Relaxation in a Superconductor

The injected spin polarized QP’s will accumulate at the interface and relax through various spin-flip mechanisms, e.g spin-flip scattering by magnetic impurities and spin-orbit interaction. We first consider relaxation only by spin-orbit scattering [64,101,102]. The created spin current density in S becomes \( J_S \approx -eD_S \nabla S \), where \( D_S = 2f(\Delta) D_N/\chi_S(T) \) is the spin diffusion constant in S and \( \chi_S(T) \) is the Yosida function [92,105]

\[
\chi_S(T) = -2 \int_{\Delta}^{\infty} D_S(E) \frac{\partial f}{\partial E} dE,
\]

which represents the reduction of the tunnel conductance by opening of the gap below \( T_C \). Furthermore, the QP population decreases and hence also their effective conductivity as the temperature is decreased. As a result, a much larger spin splitting, \( \Delta \mu \), is required in order to maintain the same spin injection and relaxation rates in S, i.e

\[
\sum_i f_i^s + e \left( \frac{\partial S}{\partial t} \right)_{sf} = 0,
\]

where \( G_{T\sigma} \) is the N-state tunnel conductance for electrons with spin \( \sigma \), \( D_S(E) \) is the normalized BCS DOS (Eq.4.11), \( f(E) \) is the Fermi function (Eq. 4.14) and

\[
S = \sum [f_{k\uparrow} - f_{k\downarrow}]
\]

is the normalized spin density in S, with \( f_{k\sigma} \) being the distribution function for QP’s in S. The spin accumulation process in S is illustrated in Figure 4.4.
where $I_i^s$ are the spin injection and extraction rates through junction $i$. Solving this equation gives the spin density $S \approx 2N_N\chi_S\Delta\mu$. Using Fermi’s golden rule [102] and the matching condition of the spin currents across the barrier, Takahashi and Maekawa [64] calculated the spin signal in a superconductor to be, 

$$R_S^{(S)} = \frac{P^2R_N}{2f(\Delta)}e^{-x/\lambda_{sf}}. \quad (4.22)$$

This expression is similar to the spin signal in N and can be obtained by scaling $\rho_N \rightarrow \rho_N/[2f(\Delta)]$. As $\Delta(T)$ increases with decreasing $T$, the spin signal increases dramatically. The spin relaxation time in S, $\tau_{sf}^{(S)}$, is determined from $(\partial S/\partial t)_{sf} = -S/\tau_{sf}$ to be

$$\tau_{sf}^{(S)}(T) = \frac{\chi_S(T)}{2f(\Delta)}\tau_{sf}, \quad (4.23)$$

which is the same result as that of Yafet [106], who studied the electron-spin resonance in S. Finally, the spin diffusion length in S is expected to be the same as in N, due to a mutual cancellation of diverging factors [64].

Zhao and Hershfield [100] and Morten et. al. [103, 104] investigated both spin-orbit and magnetic impurity induced spin flip. These two mechanisms have different Hamiltonians, and due to different time-reversal symmetry they obtain different coherence factors. As a result, the energy and temperature dependence of the spin-flip rate is different for the two mechanisms. They predict that spin-flip by magnetic impurities is enhanced for QP energies close to $\Delta$, whereas spin-flip due to spin-orbit interaction is the same as in the N state. Following the approach by Morten et. al., we assume that the spectral properties of the aluminum are given by the spatially homogeneous BCS solutions with the temperature dependence of the gap $\Delta \approx 1.76T_C\tanh(1.74\sqrt{t-1})$, where $t = T/T_C$. This assumption is valid when the contacts to the superconductor are weak and with spatial dimensions smaller than the S coherence length.

In the linear response limit, the non-local spin signal at the detector contact at a distance $x$ away from the injection point becomes

$$R_S(x) = P^2R_N \frac{g(x/\lambda_{sf}, t)}{\chi_S(t)h(t)}. \quad (4.24)$$

where $g(x/\lambda_{sf}, t)$ and $h(t)$ are rather complex energy integrals that can be approximated in the superconducting state: $h(t) \approx (1 - P^2)\chi_S(t)$ and

$$g(x/\lambda_{sf}) \approx \int \frac{\partial f(E)}{\partial E} \frac{-4N_S^2(E)e^{-x/(\lambda_{sf}\alpha)}}{2\alpha + N(E)R_N/R_I}dE.$$

Here $R_I$ is the injection tunnel resistance, $\alpha = \sqrt{(E^2 - \Delta^2)/(E^2 + \beta\Delta^2)}$ gives the renormalization of $\lambda_{sf}$ in S and $N_S(E)$ is the DOS of S. The fitting parameter
4.9. SUMMARY

\[ \beta = \frac{\tau_{so} - \tau_m}{\tau_{so} + \tau_m} \], where \( \tau_{so} \) and \( \tau_m \) are the spin-orbit and magnetic impurity spin relaxation times, respectively, is a measure of the contribution for the two scattering mechanisms. The spin flip rate in the S-state is enhanced when the dominant spin relaxation mechanism is due to magnetic impurity scattering. Equal spin relaxation in S and N is expected for spin-orbit scattering. For example, \( \beta \) is expected to approach 1 if magnetic impurities dominate spin flip processes, i.e., \( \tau_m \ll \tau_{so} \), which results in a substantial decrease in the spin diffusion length in S, \( \lambda_{sf}^{(S)} \). For dominating spin-orbit induced spin flip, i.e., \( \tau_m \gg \tau_{so} \), \( \beta = -1 \) which gives \( \alpha_l = 1 \), so that there is no renormalization of \( \lambda_{sf} \) in Eq. 4.24. The effective \( \lambda_{sf} \) can be extracted from Eq. 4.24 by an exponential model for the change in \( R_S \) between the two detectors at \( x_1 \) and \( x_2 \). The nature of spin relaxation in S is studied experimentally in paper 4 appended.

4.9 Summary

The most effective way to inject spin-polarized electrons from a ferromagnet into a superconductor is through tunnel junctions and at low injection energies, which diminishes proximity effects, Andreev reflection and charge imbalance. The spin-polarized QP’s accumulate at the interface and relax through spin-orbit interaction and/or magnetic impurities. The spin signal is greatly enhanced in the S since the QP DOS is decreased by opening of the gap. As a result a much larger spin splitting is required in order to balance the spin injection and relaxation rates in the superconductor. The spin diffusion length is equal in both states if the main relaxation process is due to spin-orbit interaction. On the other hand, it is reduced in the superconductor if magnetic impurities dominate the spin relaxation. These theoretical predictions are investigated using a multi-terminal device with a superconducting one-dimensional transport channel.
Chapter 5

The Spin Hall Effect (SHE)

5.1 Normal State

From basic physics, it is well known that a moving charge in a magnetic field will experience the Lorentz force, \( \mathbf{F} = q(\mathbf{v} \times \mathbf{B}) \). Hence, an electric current flowing in a conductor in a magnetic field will be deflected to one side of the conductor, as shown Figure 5.1 (a). The electrons will build up and cause a measurable voltage between the two sides of the conductor. Edwin Hall was the first to observe this effect back in 1879, which now bears his name, Hall effect (HE). In ferromagnets, additionally to the HE there is a contribution from the anomalous Hall effect (AHE), which is proportional to the magnetization of the F, see Figure 5.1 (b). The AHE originates from skew scattering (SS) [107, 108] and side-jump (SJ) scattering [109, 110] due to the spin-orbit interaction and impurity scattering. The SS gives the center of gravity of the incident electrons an inclined trajectory whereas the SJ gives it a transverse displacement [111]. The controversy, whether or not the AHE originates from the extrinsic SS and SJ or has an intrinsic contribution from the electronic band structure has not been completely resolved yet [107–110, 112–114]. A recent theoretical study [115] reveal that the AHE is mostly due to the extrinsic SS in clean samples, which crosses over to the intrinsic contribution for dirtier samples.

How about the case of a non-magnetic metal without any external magnetic field present? Since the spin-orbit interaction is present also in non-magnetic metals, the conduction electrons can experience similar spin-dependent scattering as in the F case [117]. Electrons moving in an electric field, i.e periodic potential from the lattice, will experience it as a magnetic field due to relativistic effects. As a result, a non-polarized spin current will undergo asymmetric scattering, i.e spin-up electrons are preferentially scattered in one direction perpendicular to the current flow, and spin-down electrons are preferentially scattered in the opposite direction, as illustrated in Figure 5.1 (c). The direction is determined by the cross
CHAPTER 5. THE SPIN HALL EFFECT (SHE)

Figure 5.1: Different Hall effects. (a) Ordinary Hall effect (HE): the charge carriers moving in a magnetic field experience the Lorentz force and are deflected to one side, creating a transverse Hall voltage but no spin accumulation. (b) Anomalous Hall effect (AHE): results from a spin-dependent scattering in ferromagnets and adds to the ordinary HE. This causes both a Hall voltage and a spin accumulation. (c) Spin Hall effect (SHE): results from a spin-dependent scattering in non-magnetic conductors due to the spin-orbit interaction. (d) An un-polarized current will be equally deflected to each side resulting in a zero transverse spin Hall voltage, $V_{SH} = 0$. (e) A spin-polarized current, produced by e.g. spin injection, will result in a different number of electrons on each side and thus creates a finite voltage $V_{SH} \neq 0$. (d-e) after [116].

The product [118–120]

$$\text{direction} \sim \hat{\sigma} \times \hat{k},$$

(5.1)

where $\hat{\sigma}$ denotes the spin index and $\hat{k}$ represents the direction of the electron current. This separation of spin is referred to as the direct spin Hall effect (SHE), and depends on the strength of the spin-orbit interaction in the material. Accordingly, the conversion of spin current into a charge current is referred to as the inverse SHE, and is consistent with the Onsager symmetry relation. Since the initial spin current is un-polarized, equal number of electrons end up at each of the two sides of the sample, resulting in a zero transverse voltage, Figure 5.1 (d). However, if the
current is initially spin-polarized, by e.g. spin injection from a F, different number of electrons will travel to either side of the sample, and finite voltage ($V_{SH}$) arises, as shown in Figure 5.1 (e). Observe that the magnetization direction in this case must be put out of the plane in order for the electrons to scatter to the left or right.

The SHE was first predicted in 1971 by D'yakonov and Perel [121, 122], followed by the theoretical study on impurity scattering by Hirsch [117] and an extension of the theory to the diffusive transport regime [118]. The first experimental observation of the direct SHE was achieved via the magneto-optical Kerr effect in a GaAs semiconductor channel [123]. Using similar optical measurement technique, the intrinsic SHE was observed in a 2 DEG system [124,125]. The advantage of using semiconductors is the extremely long $\lambda_{sf}$ which makes the detection by a laser beam possible. The reason is that the spot size ($\sim 1\,\mu m$) of the laser beam is smaller than $\lambda_{sf}$. This can not be applied in metal films, where $\lambda_{sf}$ is typically shorter than $1\,\mu m$. The electrical detection of the SHE in semiconductors is still to be demonstrated.

Several experimental configurations have been proposed with the aim to electrically measure the SHE in normal metals [117–120] and in superconductors [126]. However, these scheme are difficult to implement due to limitations in fabrication. Also, spurious effects such as the anisotropic magneto resistance (AMR) effect, spin-flip scattering at interfaces and other Hall effects in the ferromagnets could washout the wanted signal. By using the non-local spin injection technique discussed in detail above these effects are avoided and the spin dependent signals isolated. In fact, very recent experiments employ this to demonstrate the inverse SHE in a lateral CoFe/Al$_2$O$_3$/Al device at 4 K [16] in a measurement setup equivalent to that illustrated in Figure 5.1 (e). Aluminum is known to have a weak spin-orbit interaction since it is a light metal. Using a material with a much stronger spin-orbit coupling (Pt), both the direct and the inverse SHE, at room temperature and at 77 K, were demonstrated [19]. Nevertheless, the measured signals are very weak, $\sim 10\,nV$. We have studied this effect in the superconducting state, which is discussed below and in more detail in paper 5 appended.

5.2 Superconducting State

As mentioned earlier, a superconductor is very sensitive to magnetic fields and high current densities. For example, the strong out of plane magnetic field, needed to saturate the magnetization direction vertically [16] would completely destroy the superconductivity. Also, the high QP currents needed to create large enough spin/charge currents [19] would suppress the superconductivity [99]. Thus, we need a different kind of device in order to demonstrate the SHE in a superconductor.

The new design is illustrated in Figure 5.2, where several F electrodes act as QP injector and detectors via tunnel barriers. The use of tunnel barriers is crucial since it diminishes the parasitic influence of the proximity effect and/or Andreev
Figure 5.2: Illustration of the experiment, where a ferromagnetic electrode is used to inject spin-polarized quasi-particles (QP’s) via a tunnel barrier into a superconducting wire. Due to the short spin diffusion length in the wire, the QP’s are completely un-polarized as they reach the detectors. At this point, spin-up and spin-down QP’s will be separated in the $\hat{y}$-direction due to the spin Hall effect (SHE). Hence, opposite spin accumulations are created at the two surfaces of the wire.

processes [80]. It also provides a uniform current injection [13] and enhances the spin-polarization [15, 127]. From the left F electrode, spin polarized electrons are injected into the superconducting Al wire and diffuse exponentially with the characteristic time (length) scale $\tau_{sf} (\lambda_{sf})$. The injected electrons remain unpaired QP’s for a long time ($\tau_r$) compared to that of spin relaxation ($\tau_{sf}$), before they combine into Cooper pairs and condense in the superconducting state, i.e $\tau_r \gg \tau_{sf}$ [83, 84]. In fact, it will be shown that $\lambda_{sf}$ in S is short enough for the spin of the diffusing QP’s to be completely randomized already after $\sim 100$nm. Thus, the spin accumulation is zero at $x >> \lambda_{sf}$ and only un-polarized QP’s reach the detectors. The QP current is separated into two spin components due to the SHE, where the spin-up (+$\hat{z}$) QP’s will be accumulated at the top Al surface and the spin-down ($-\hat{z}$) will be accumulated at the bottom Al surface, according to the cross product in Eq. 5.1. Detector 1 and 2 are spin sensitive, which enable us to measure the amount of spin accumulated at the top surface, and thus gives us the opportunity to determine the SHE in the system.

We follow the theoretical approach by Takahsahi and Maekawa [126, 128] in describing the SHE in our experiment. The diffusing QP current $[j_Q(x)]$ creates a transverse spin current $[J_{SH}(x)]$ in the $\hat{y}$ direction, due to the direct SHE, i.e SS and SJ scattering, and is given by

$$J_{SH} = \left[ \eta_{HI}^{SJ} + \frac{\lambda_Q}{2f_0(\Delta)} \eta_{HI}^{SS} \right] j_Q - 2f_0(\Delta)\sigma_N \nabla_y \delta \mu_{SH},$$

(5.2)

where $\eta_{HI}^{SS}$ and $\eta_{HI}^{SJ}$ are the SS and SJ coupling parameters, $f_0(\Delta) = 1/(\exp(\Delta/kT)+1)$ is the Fermi distribution function, $\sigma_N$ is the N-state conductivity, $\delta \mu_{SH}$ is the
5.2. SUPERCONDUCTING STATE

Figure 5.3: (a) Scanning electron microscopy (SEM) image of the device. The distance from the injector to detector 1 and 2 is $x_1 = 900$ nm and $x_2 = 1200$ nm, respectively. (b) Spatial dependence of the spin accumulation at the surface ($\delta \mu_{SH}$) created by the deflection of the QP current due to the SHE. The spin-sensitive detectors 1 and 2 enable us to measure this spin accumulation.

Spin accumulation in $y$ and $\chi^0_Q$ is the QP susceptibility

$$\chi^0_Q(T) = 2 \int_0^\infty \frac{\sqrt{E^2 - \Delta^2}}{E} \left( - \frac{\partial f_0(E)}{\partial E} \right).$$

The induced spin accumulation at the surfaces, decays exponentially with decay-
ing QP current, according to

\[ \delta \mu_{SH}(x) = \frac{e \rho_N d}{2 f_0(\Delta)} \left[ \eta_{HJ}^S + \frac{\chi_Q^0}{2 f_0(\Delta)} \eta_{HH}^S \right] j_Q(0) e^{-x/\lambda_Q}, \tag{5.4} \]

where \( \rho_N \) is the N state resistivity, \( d \) is the wire thickness and \( \lambda_Q \) is the average distance traversed by the QP’s before recombining into Cooper pairs, i.e recombination length. The sample is designed such that \( x_1, x_2, (x_2 - x_1) \ll \lambda_Q \), thus avoiding the contribution from charge diffusion.

Figure 5.3 (b) shows the spatial dependence of \( \delta \mu_{SH} \) and illustrates the voltages measured with the detectors in the P and AP magnetic configurations. The blue curve corresponds to the detection of spin-up (P) electrons and the red curve corresponds to the detection of spin-down (AP) electrons. Hence, by putting the detectors in the AP state, both spin directions can be measured. Depending on the magnetic states of the detectors, the voltage difference will be either low, in the P state, or high, in the AP state. The SHE resistance differentially measured by the detectors is \( R_{SH}(x) = (V_1 - V_2) / (A j_Q(0)) \), where \( P \) is the effective polarization and \( A \) is the junction area. Moreover, the difference in the resistance between the AP and P state is defined as the SHE signal, \( \Delta R_{SH} = (R_{SH}^{AP} - R_{SH}^{P}) \), and is given by

\[ \Delta R_{SH} = \frac{P \rho_N}{w 2 f_0(\Delta)} \left[ \eta_{HJ}^J + \frac{\chi_Q^0}{2 f_0(\Delta)} \eta_{HH}^S \right] \left( e^{-x_1/\lambda_Q} + e^{-x_2/\lambda_Q} \right). \tag{5.5} \]

The reason that we take the difference in resistances, is to rule out any offsets created by the inductive pick-up from the electromagnetic environment. Also, note that the distance between the detectors (~300 nm) is much smaller than the recombination length \( \lambda_Q \approx 5 - 10 \mu m \) [129], and thus effects of the QP recombination can be neglected. The measurements are discussed in chapter Results and paper 5 appended.
Chapter 6

Experimental Methods

6.1 Introduction

Since spin transport occurs only on sub micron scales the structures must have very small dimensions in order to observe any spin dependent effects. We produce the suitable nano structures by exposing double layer resist masks using an electron-beam lithography (EBL) system and thereafter e-gun depositing desired materials. It must be mentioned that this part of this PhD study was the most time consuming. Due to the small dimensions of the samples they become very delicate and sensitive to external influences such as voltage spikes, dirt, handling, etc.

Measurement wise, the aim was to detect low voltages, < 100 nV, with good enough accuracy. Different techniques were employed for this purpose, where the most common one was the standard lock-in technique with specially designed small signal, pre-amplifiers.

6.2 Sample Fabrication

Electron beam lithography (EBL)

All samples were prepared on pre-oxidized silicon wafers with approximately 1 μm of SiO₂. Two layers of resist with different thicknesses and different sensitivity for e-beam exposure are spun on to the wafers. The bottom layer consists of PMGI SF7 which was spun at a speed of 3000 rpm for 1 min and then baked for 10 min at 180°C to give a thickness of 400 nm. The top layer consisting of ZEP 520, a high resolution e-beam resist, diluted 1:2 in Anisole was fabricated in the same way but was spun with twice the speed to give a nominal thickness of 70 nm.

The EBL system used for exposure of the samples is a Raith 150. It consists of a scanning electron microscope (SEM) from LEO with an additional pattern generator, a high-precision laser-interferometric stage and a beam blanker from RAITH GmbH. The Gemini column provides an accelerating voltage of 30 kV and
an electron beam diameter as small as 1.2 nm at the specimen. The samples were exposed in two steps, where the first step contained the smallest structure and was exposed with the smallest aperture size of 7.5 μm and beam current of ≈ 20 pA. The second pattern contains the connecting leads to the small structure and the bonding pads. The pads are much larger and do not need to be as well defined as the smallest structure. Hence the largest aperture size of 120 μm with a beam current of ≈ 5 nA is used, which significantly reduces the exposure time.

The long copolymer molecules in the resists are broken into shorter molecules when exposed to electrons and then dissolved by chemical solvents. The top layer is developed in p-Xylene for 70 s to dissolve the exposed ZEP 520 areas. Afterwards, the chips are dried with nitrogen and then put in diluted MF322 solvent (3:2, MF322:H₂O) for 95 s to dissolve the exposed bottom layer only. This layer has higher exposure sensitivity, which makes it possible to create undercuts. Such process allows to obtain a mask of ZEP 520 with free standing bridges, supported at both ends by remaining PMGI SF7. The interplay between the distance of the structures (≈ 150 nm), the dose, development time and chemical mixture determines the exposed areas undercut depth. This is crucial in order to obtain an overlap between different electrodes. Finally, the chips are rinsed in de-ionized water and dried again in nitrogen. The process is illustrated in Figure 6.1. The chips are now ready for material deposition.

Figure 6.1: Sample fabrication: (1) spinning of double layer resist (2) EBL exposure of the structure (3) development of the exposed areas (4) deposition of Al (5) oxidation of Al to obtain tunnel barriers (6) deposition of Co at an angle to overlap the Al.
Electron-gun evaporation

Co/Al tunnel junctions are fabricated using shadow evaporation technique, [130]. First, a 100 nm wide Al strip of various thickness is deposited, onto the Silicon wafer. Next, without breaking the vacuum, the Al strip is oxidized in pure oxygen for 10-20 min at a pressure of 0.1-1 mBar. A thin oxide layer builds up at the surface serving as a tunnel barrier with a typical junction resistance of 30-80 kΩ. Moreover, the oxidized Al on the resist mask strengthens the free standing bridges and thereby prevents a mask deformation during the subsequent Co evaporation.

The second evaporation step consists of Co deposited at an angle of 40° in order to overlap the Al strip. Since the Co can heat and deform the mask, it was deposited in several steps with 5 min of waiting in between. The Co electrodes were made with different widths, 50-80 nm, in order to achieve different coercivities and thus making it possible to switch the magnetization direction of the individual electrodes. Finally, the remaining resist was lifted off by a solvent (Microposi Remover 1165, Shipley) at 55°C, leaving on the substrate only the metallic nano-structure. The complete process is illustrated in Figure 6.1 and 6.2. Figure 6.3 shows SEM images of a fabricated sample at different magnifications. In the top figure, the entire structure (1.5×1.5 mm) is visible, from bonding pads to connecting leads. Magnifying 50 000 times, reveals the actual Co electrodes which overlap the Al strip. This multi terminal geometry was chosen in such a way as to enable us to perform different kinds of experiments on the same chip.

Figure 6.2: Three dimensional illustration of the deposition process.
Figure 6.3: SEM pictures of a sample at different magnifications. (a) A cage with 15 bonding pads. (b) Zoom of the small box, of about 1000 times. (c) After a magnification of $50 \cdot 10^3$ times, a few of the many electrodes become visible. The Co electrodes (50-70 nm thick) overlap the oxidized Al strip (15-35 nm thick) and form tunnel junctions. The Co electrodes have different widths in order to achieve different coercivities.

### 6.3 Sample mount and wires

The samples were glued on to two kinds of chip carriers, one with 5 and one with 8 twisted pair cables, respectively, as shown in Figure 6.4. The left chip carrier is a specially designed PC-board for our dilution refrigerator. The right one is a standard 16-way dual-in-line chip carrier and is used in $He^4$ and $He^3$ cryostats. The pads on the sample and those of the chip carriers were inter bonded with 25 μm Al wire using a Kulicke and Sofa wedge-wedge bonder. To further connect the chip carrier with the measurement setup, it is mounted on a DIL socket with twisted pairs connected to its pins. The advantage of using twisted pairs is that it reduces the electro magnetic noise pick-up from the environment since it minimizes the loop area between the two cables. It also confines parts of the electric and magnetic fields generated by signals in the cables to within the spiral, preventing the cable to act as an antenna.
6.4 COOLING DOWN

Figure 6.4: (a) Specially designed PC-board for our dilution refrigerator, with 5 twisted pair cables. (b) Standard 16-way chip carrier for dipsticks and a $^3$He cryostat.

In our case, the twisted pair cables are especially useful since we are measuring differential voltages, i.e one cable for "high" and one for "low" (ground) signal. Provided that the cables have equal impedance, any electrical noise pickup will be equal, i.e flowing in the same direction with equal amplitude and hence resulting in equal voltages in each cable. The differential pre-amplifier will sense this as a common mode signal and will reject it. It is therefore important that the cables are as identical as possible. They are made of Constantan which has a rather constant resistance (65 $\Omega$/m) against temperature.

6.4 Cooling Down

Several cryostats were used to cool down the samples. The simplest and most effective one is a home built flow cryostat which reaches temperatures down to 2 K. Lower temperatures are achieved by the use of Oxford Instrument Heliox$^L$ sorption pumped $^3$He cryostat. The temperature control of this cryostat enables us to do experiments in the range 0.26 - 100 K. This was necessary when studying the temperature dependence of the spin dependent properties. Ultra low temperatures are obtained by an Oxford Kelvinox AST Minisorb dilution refrigerator with a base temperature of 14 mK and a cooling power of roughly 40 $\mu$W at 100 mK. To maintain the base temperature it is important to have a good mechanical contact between the sample and the cooling part of the cryostat, i.e the mixing chamber. Therefore, the chip is tightly placed in a radiation tight copper enclosure, thermalizing it well with the copper box, and then screwed to the mixing chamber. Inside the dewar, a superconducting (Nb-alloy wire) electro magnet is positioned with a diameter large enough to fit the cryostat. The electromagnet is driven by a Yokogawa voltage source together with a bipolar power amplifier, kepco BOP400. This way we achieve an in-plane magnetic field which gives us the possibility to switch the magnetization direction of the electrodes.
6.5 Measurements

We are interested in measuring very small voltages, < 100 nV, with an accuracy of \( \sim 1 \) nV. Such small voltages are often in the same order or below the noise. It is therefore essential to minimize the factors contributing to noise. This is realized by using clean samples, short cables, low noise pre-amplifiers, RC-filters, good shielding, avoiding ground loops etc. Several types of electrical measurements were performed: conventional DC I-V measurements, non-local DC I-V measurements and non-local AC and AC+DC measurements. The typical measurement setup is illustrated in Figure 6.5, where current or voltage is biased through the right electrode and non-local voltages are measured in the left electrodes. The voltages were amplified by high input impedance instrumentational pre-amplifiers, INA 116, with a typical input bias current of 3 fA. They consisted of two Burr-Brown OPA111BM at the inputs, connected to a Burr-Brown differential amplifier INA 105KP. The last stage had a variable gain of 1, 10, 100 and 1000.

For the DC measurements an HP function generator was used to sweep a triangular wave with frequency of 0.125 Hz. The voltages were pre-amplified 1000 times and sampled in a 4 channel Agilent (Infiniium) Oscilloscope. A total of 225 points were sampled during each sweep (period) where the last 25 points were used to dynamically compensate for the offset in the signal, i.e. auto zeroing. During this time of the period, 8 s, the drift of the amplifiers were negligible. Using

![Figure 6.5](image_url)

Figure 6.5: Non-local measurement setup, i.e voltages are measured outside the current path. AC, DC and AC+DC measurements are performed while sweeping external magnetic field, \( H \), which is needed for switching the magnetic states of the electrodes.
6.5. MEASUREMENTS

This style of measurement enables us to characterize the samples in terms of resistivity, tunnel barriers, critical current and superconducting gap. The non-local measurement setup is especially useful for bias dependence measurements.

The drawback with DC measurements is the higher noise level compared to the lock-in technique. Lock-in amplifiers are used to measure the amplitude and phase of signals buried in noise. This is achieved by allowing only signals of a certain frequency to pass while the unwanted noise is removed, i.e., using the lock-in amplifier acts as a narrow bandpass filter. The frequency of the signal to be measured and hence the passband region of the filter is set by a reference signal, which has the same frequency as the modulation of the signal to be measured. An additional advantage of the lock-in technique is that it eliminates amplifier drifts.

The samples were AC biased at frequencies of 1-10 Hz through a large resistor of typically 1 MΩ which was connected in series between the output of the lock-in and the sample to give a bias current of 1 μA. The signals were pre-amplified 1000 times and then phase sensitively measured using lock-in voltmeters, SRS 830. Using long time constants we were able to obtain the desired sensitivity of 1-5 nV.

Another measuring technique is to apply an AC signal superimposed on a stepped DC bias and then use a lock-in amplifier to obtain the AC voltage across and the AC current through the sample. This is equivalent to performing differential conductance measurement. A Keithley 6221 current sources is used for this purpose which combines the DC and AC components into one source, with no need to do a secondary measure of the current. This technique is also useful for bias dependence measurements since the DC component can be swept as desired. Another advantage is that the voltages are measured with sensitive lock-in amplifiers.

All these measurements were done in different magnetic configurations of the Co electrodes, i.e., parallel (P) or anti-parallel (AP). The system was put into either magnetic state by an external magnetic field directed along the electrodes. The field is then turned off and the desired measurements were conducted. This procedure is especially necessary in the superconducting state since the external magnetic field can suppress superconductivity.
Chapter 7

Results in the Normal State

7.1 Spin Injection and Relaxation in Normal Metal Nanowire

This chapter outlines the main results of our experiments. Our multi-electrode device, discussed throughout the thesis (Figure 6.3), allows a direct measurement of the spin accumulation ($R_S$) and spin diffusion length ($\lambda_{sf}$). It is therefore a perfect tool for exploring the fundamental properties of spin transport in metallic nanowires. The main scattering mechanisms are determined by studying the thickness and temperature dependence of the resistivity.

7.2 Surface Scattering

Several mechanisms contributing to the resistivity in thin films were discussed in chapter Thin film resistivity. Considering the physical parameters included in modeling these mechanisms, the Soffer-Mayadas-Shatzkes model (Eq. 2.7) was argued to give the most adequate physical picture in our case. To confirm this, we studied the thickness dependence of the resistivity and hence the contribution to it from surface scattering. An atomic force microscope (AFM) was used to measure the average grain size $D$ and surface roughness for the different films (Figure 7.1 (a)). $D$ did not change with thickness and was estimated to be $D \approx 7$ nm throughout the thickness range studied. The surface roughness was also approximately constant, $\approx 1$ nm r.m.s.

Figure 7.1 (b) shows the resistivity of Al films measured at 4 K as a function of thickness together with a theoretical fit according to Eq 2.7. The measurements were performed at low temperatures in order to reduce the electron-phonon scattering. The fact that $D$ essentially did not change with thickness, results in approximately a constant contribution to the total resistivity from grain boundary scattering. This is represented by the dashed line, as a pure grain boundary scattering assuming ideal surface specularity ($p_i = 1$). If the contribution from surface scattering is taken into account, the theory agrees with our experimental data per-
CHAPTER 7. RESULTS IN THE NORMAL STATE

Figure 7.1: (a) AFM images of a 40 nm thick Al film. Average grain size $D \approx 7$ nm and the r.m.s roughness $\approx 1$ nm. (b) Resistivity of Al films as a function of thickness together with theoretical fit using Eq. 2.7. The solid line represents $\rho_f$ where both grain boundary and surface scattering is taken into account. The dashed line represents $\rho_f$ only due to grain boundary scattering and assuming ideal surface specularity.

fectly. The solid line represents the theoretical prediction for diffusive surfaces and $D \approx 7$ nm. Thus, the dominating contribution to the resistivity of our thin films is the diffusive ($p_i = 0$) scattering at the surfaces, such as in [12, 13, 131].

7.3 Spin Relaxation due to Surface Scattering

In this subsection, the mechanisms for spin relaxation in the N-state will be discussed. In particular spin-flip scattering at the surfaces. The measurements were performed using the lock-in technique, with a bias current of $I_{bias} = 5 \mu A$ (see chapter Measurements for details). Figure 7.2 (a) and (c) show two measurement configurations for two samples with different thickness, both using the non-local measurement technique. Current is biased through the injector and the non-local voltages are measured at 300 nm and 600 nm from the injector, using two other electrodes, detector 1 ($D_1$) and detector 2 ($D_2$), respectively. Figure 7.2 (b) shows $R_s = V/I$, measured individually, as a function of applied magnetic field, $H$, at 4
Figure 7.2: Non-local measurements for two samples of different thickness, performed at $T = 4$ K. The curves have been shifted vertically for comparison purposes. The arrows indicate the relative magnetization direction of the electrodes. 

(a) Measurement setup, with individual non-local detection. (b) Spin signals measured with detector 1 ($D_1$) at 300 nm (solid line) and with detector 2 ($D_2$) at 600 nm (dashed line) as a function of applied magnetic field. (c) Measurement setup, with a differential non-local detection. (d) Differential spin signal as a function of applied magnetic field.

K. To begin with, the magnetization of the electrodes is saturated in the negative direction (↓↓↓) and then switched separately by ramping the field in the positive direction. First, $D_1$ switches (↓↑↓) at 1.2 kOe, then $D_2$ (↑↑↓) at 1.7 kOe and finally the injector (↑↑↑) at 1.8 kOe, which can be seen as a simultaneous switch in both curves. Figure 7.2 (d) shows the differential spin signal, $\Delta R_{S12} = \Delta V_{12}/I$, as a function of $H$, which is an equivalent way of obtaining $R_{S1}$ and $R_{S2}$. Note that the actual spin signal is the difference between the P and the AP-state, labeled as $R_{S1}$ and $R_{S2}$.

At 4 K, the typical junction resistances were 50-200 kΩ and the Al resistivity 2-5 $\mu\Omega$cm. Using the Einstein relation $\sigma = e^2 N_{Al} D_N$, with $N_{Al} = 2.4 \times 10^{28}$ eV$^{-1}$m$^3$ [13] being the density of states at the Fermi level, gives the diffusion con-
const $D_N = (3 - 9) \times 10^{-3}$ m$^2$s$^{-1}$. Now, fitting the data from Figure 7.2 (b) and (d) to Eq. 3.30 yields $\lambda_{sf} \approx 1000$ nm, $\tau_{sf} \approx 100$ ps and the effective spin polarization of $P = 12\%$. Note that these spin transport parameters are obtained \textit{in-situ} in a single field sweep which eliminates uncertainties due to irreproducibilities in fabrication. They are in good agreement with the recent results for similar structures [13–16].

The above procedure is repeated for several samples with different thicknesses, in order to investigate the contribution from the surfaces to spin relaxation. As the thickness of our samples is decreased, the resistivity is greatly enhanced, i.e the momentum mean free path ($\lambda_e$) and time ($\tau_e$) of the electrons are strongly reduced, as shown in Figure 7.3, indicating that the scattering from the surfaces determines $\lambda_e$. If the spin relaxation is governed by the same mechanisms, $\tau_{sf}$ and $\lambda_{sf}$ should follow the same behavior as $\tau_e$ and $\lambda_e$. In fact, the ratio between them should be constant, which obviously is not the case in our samples. Figure 7.3 (a) shows that $\tau_e$ decreases with thickness whereas $\tau_{sf}$ remains almost constant, $\tau_{sf} \approx 100$ nm. This means that the surface scattering contribution to spin relaxation is weak compared to that within the bulk of the Al films. This is also illustrated in Figure 7.3 (b), where $\lambda_{sf}/\lambda_e$ is plotted, clearly showing that the ratio is not constant.

The observed significantly reduced spin flip scattering at the surfaces compared to that in the bulk can be expected from at least two mechanisms [132]. First, the spin hot spots discussed in section \textit{Mechanisms of Spin Relaxation}, which are due to a peculiar band structure of Al and which dominate the intrinsic spin flip

![Figure 7.3](image-url)

Figure 7.3: Thickness dependence of the spin relaxation parameters, illustrating the contribution from surfaces. (a) Thickness dependence of the spin relaxation time ($\tau_{sf}$) and the momentum mean free time ($\tau_e$) at 4 K. For easier comparison, $\tau_e$ is multiplied by a factor of $10^4$. (b) Ratio between the spin diffusion length ($\lambda_{sf}$) and the momentum mean free path ($\lambda_e$) as a function of Al film thickness at 4 K. The line is a guide for the eye.
scattering in the material, are expected to be affected by the presence of the interface where the band structure is significantly altered. Secondly, the nature of impurities at the interfaces can differ from that in the bulk of Al. One can expect the fully or partially oxidized Al surfaces to have some amount of charge impurities (so-called random background charges in oxides), which affect the conduction through the Coulomb interaction but are unable to flip spin. In the bulk, on the other hand, some presence of magnetic impurities could be the channel for both momentum and spin flip scattering.

7.4 Spin Relaxation due to Impurity Scattering

In addition to scattering from surfaces for very thin films, there is always background impurity scattering within the material. To investigate how this contributes to the spin relaxation, the temperature dependence of $\tau_{sf}$ is determined, see Figure 7.4. At higher temperatures ($T > 50$ K), the phonon-mediated scattering enhances the spin-flip scattering. As $T$ is lowered, $\tau_{sf}$ increases from $\approx 50$ ps at room temperature until it levels off at $\approx 120$ ps at 4 K. This indicates that the spin relaxation is impurity dominated at low temperatures. Figure 7.4 shows the experimental data on $\tau_{sf}$ for single crystal Al by Johnson and Silsbee [9, 11] and Lubzens and Schultz [133], as well as for thin Al films by Jedema et. al. [13] and us. In our case the thickness of the film is 15 nm.

![Figure 7.4](image)

Figure 7.4: Temperature dependence of the spin relaxation time ($\tau_{sf}$), for different research groups, together with a theoretical fit according to Fabian and Das Sarma [76].
The total spin flip scattering can be separated according to the Matthiessen’s rule [134],

\[
\frac{1}{\tau_{sf}} \approx \frac{1}{\tau_{imp.}} + \frac{1}{\tau_{ph.}},
\]

(7.1)

where \(\tau_{imp.}\) and \(\tau_{ph.}\) are the spin relaxation times due to impurity and phonon scattering, respectively. Using Fabian and Das Sarma’s model [76] in combination with Eq. 7.1 it is possible to calculate the total spin flip time \(\tau_{sf}\). The fit is illustrated as a solid line in Figure 7.4, and agrees well with our experimental data. Our results on the temperature dependence can be reconciled with the early single-crystal data [9], and the sole difference appears to be the greater amount of impurities in thin films in our case, which results in a two orders of magnitude longer \(\tau_{sf}\).

7.5 Decoupling of Spin and Charge Currents in Nanowires

The fundamental assumption that spin diffusion is isotropic has been known since the pioneering work by Johnson and Silsbee [9–11], but no direct experimental evidence has been reported. Below, we describe our direct demonstration of the strictly isotropic nature of spin diffusion in 1D transport channels.

We follow the approach described in section Symmetric Spin Diffusion, where current is sent from the injector to the right side of the Al strip, and \(R_S\) is measured with several detectors along the Al strip, (see Figure 7.5 (a) and (c)). This way, both the local \((D_1)\) and the non-local \((D_2, D_3)\) spin accumulation is probed. Since the detectors are identical and placed at equal distances from the injector, they should detect the same amount of spin accumulation. In other words, measuring the differential spin signal between \(D_1\) and \(D_2\), i.e \(\Delta R_{S12} = \Delta V_{12}/I\), should yield zero signal when the system is symmetric, i.e \((↑↑↑), (↑↑↓), (↑↓↑)\) and \((↓↓↓)\). The arrows indicate the magnetic configuration of the electrodes. On the other hand, when the system is anti-symmetric, i.e \((↑↑↑), (↑↑↓), (↑↓↓)\) and \((↓↓↑)\), the signal should be high and correspond to \(R_S(300 \text{ nm})\). This is confirmed by the data in Figure 7.5 (b) which shows \(\Delta R_{S12}\) as a function of \(H\), with the constant spin-independent resistive offset subtracted for a more straightforward comparison of the spin-dependent signals.

Initially, the magnetization of the electrodes is saturated in the positive direction \((↑↑↑)\), and thereafter switched individually by ramping the field in the negative direction. When \(D_2\) switches \((↑↑↓)\) at -650 Oe, it becomes AP with respect to the injector and thus detects a high \(R_S\), whereas \(D_2\) is still in the P-state and measures zero \(R_S\). Consequently, the system will be anti-symmetric and result in a high \(\Delta R_{S12} (≈ 275 \text{ mΩ})\), which is observed as a sudden jump in Figure 7.5 (b). Next to switch is \(D_1\) \((↑↑↓)\) at -850 Oe, which puts it in the AP-state with respect to the injector. This makes the system symmetric again, and the resulting \(\Delta R_{S12}\) is zero. In order to establish when the injector switches, simultaneous non-local measurements between \(D_2\) and \(D_3\) are performed. Figure 7.5 (d) shows \(\Delta R_{S23}\) as
7.6 SPIN ABSORPTION BY FERROMAGNETS

(a) Local (Detector 1) and non-local (Detector 2) measurement setup. (b) Differential $R_S$ between the local and the non-local detector, both at the distance of 300 nm from the injector. (c) Measurement setup, with a differential non-local detection. (d) Differential $R_S$, between detector 2 (300 nm) and 3 (600 nm).

Figure 7.5: Different measurements of $R_S$, at $T = 4$ K, demonstrating the concept of spin and charge current decoupling. The curves have been shifted vertically for comparison purposes. The arrows indicate the relative magnetization direction of the electrodes. (a) Local (Detector 1) and non-local (Detector 2) measurement setup. (b) Differential $R_S$ between the local and the non-local detector, both at the distance of 300 nm from the injector. (c) Measurement setup, with a differential non-local detection. (d) Differential $R_S$, between detector 2 (300 nm) and 3 (600 nm).

a function of $H$, where the switching of the injector (↓↓↓) is marked with a dashed line at -930 Oe. This leaves the system symmetric and therefore $\Delta R_{S12} = 0$. Moreover, this enable us to in-situ measure $R_S$ and $\lambda_s$. Thus, in a single field sweep we obtain the spin dependent transport properties of the sample and directly demonstrate that the spin and charge currents are decoupled. Note that, the behavior in the positive direction is identical.

7.6 Spin absorption by ferromagnets

Possible source of uncertainty in all-metallic nano-devices is a non-uniform current distribution over the contacts. Furthermore, as discussed in section Spin Ac-
cumulation, some of the spin current is absorbed back into F, thus diminishing
the spin accumulation. Kimura et. al. [69,135] showed that, placing an extra F
electrode between the injector and the detector absorbs spin. We observe very
similar effects, when replacing the tunnel barriers with metallic contacts. Figure
7.6 (a) shows a measurement configuration, where two identical detectors ($D_1$ and
$D_2$), are placed in tunnel contact at equal distances of 600 nm about the injector.
An additional F electrode is directly connected to the Al wire between the injec-
tor and $D_1$, acting as a spin absorber. On its way to $D_1$, a significant portion
of the spin population is lost in the absorber. This is clearly observed in Figure 7.6
(b), where $R_{S1} = V_1/I$ (dashed line) and $R_{S2} = V_2/I$ (solid line) are plotted as a
function of $H$. Owing to spin absorption, the unperturbed spin signal $R_{S1} \approx 0.22$
m$\Omega$ is reduced to $R_{S2} \approx 0.05$ m$\Omega$, which is in good agreement with the findings
of [69,135]. Measurements using tunnel barriers are non-disruptive and allow an
accurate determination of the spin sinking effect, in this case in-situ in the same de-
vice and field sweep. This result for our Co/Al interfaces differs from the Py/Ag
data on spin-sinking recently reported by Godfrey and Johnson [67]. We have not
analyzed our structure in terms of contact resistance. However, the experimental
evidence is clear that the spin absorber electrode is effective, without showing any
additional switching in the magneto resistance of Figure 7.6 (b), dashed line.

![Figure 7.6](image)

Figure 7.6: Measurements of $R_S$, at $T = 4$ K, demonstrating the concept of spin
absorption. The curves have been shifted vertically for comparison purposes. The
arrows indicate the relative magnetization direction of the electrodes. Owing to
the metallic contact F(Absorber)/Al, spins can be absorbed on the way to Detector
1. (a) Detector 1 (2) measures $R_S$ in the presence (absence) of spin absorption. (b)
$R_S(600 \text{ nm})$ measured with Detector 1 (dashed line) and with Detector 2 (solid
line).
Chapter 8

Results in the Superconducting State

8.1 Characterization of the samples

In the previous chapter we showed a straightforward and efficient approach for determining the spin dependent transport properties of a nanowire. However, it is not directly applicable in the S-state, since the high currents and magnetizing fields normally used in the N-state, destroy superconductivity. Special care must be taken not to suppress the gap in the S nanowire, where even the fringing fields from the F electrodes are known to be strong enough to affect superconductivity [136–138]. It is therefore important to verify that the spin channel is maintained superconducting throughout the magneto-transport measurements.

The fringing fields through the Al wire are drastically reduced by shifting the ends of the F electrodes by ~150 nm past the Al strip. To verify that the superconducting properties of the Al wire are not affected by the switching of the F electrodes between the P and AP configurations, $I_C$ of the wire is measured for the various magnetization states of the electrodes. The distribution of the fringing fields near the ends of any neighboring electrode pair changes significantly on P/AP switching. This should affect the superconductivity and thereby change $I_C$, only if a significant portion of this fringing field penetrated the Al wire. The electrodes are put in specific, well defined magnetic states by stopping the field sweep at the appropriate field, and returning to zero field to perform the measurement. This procedure avoids uncertainties due to a possible direct influence of the external field on S [139]. Figure 8.1 (a) shows the measurement setup for determining $I_C$ of S for different magnetic configurations of the F electrodes. Current is sent through the Al strip and voltage is measured between two F electrodes that are separated by 300 nm. The $I$-$V$ characteristics of such measurements, shown in Figure 8.1 (b), reveal that $I_C \approx 700$ nA at 250 mK, and does not change between the P and AP configurations. This means that the possible changes in fringing fields between P/AP switching do not alter the superconducting parameters relevant for spin transport.
Figure 8.1: I-V curves for different current paths and magnetic configurations, measured at 250 mK. (a) Measurement setup, where current is sent through the Al strip and voltage is measured between two F electrodes, separated by 300 nm. (b) I-V curves for the setup in (a) for the P (straight line) and AP (circles) configurations. (c) Measurement setup, where current is sent through the tunnel junctions and voltage is measured between the ends of the Al strip. (d) I-V curves for the setup in (c) for the P (straight line) and AP (circles) configurations.

It is known, that injection of high energetic QP’s suppress superconductivity due to the pair breaking effect [78, 79, 99]. To investigate how this affects our superconducting nanowire, we now inject electrons through the tunnel junctions, as shown in Figure 8.1 (c). This way, QP’s are created and injected into S, and the voltage is measured between the ends of the Al strip. A negative influence on the superconductivity would manifest as voltage change. In fact, $I_C$ in this setup drops by a factor of 4, as shown in Figure 8.1 (d), compared to that in (a). Nevertheless, it remains unchanged between the different magnetic configurations. In the P-state, the same amount of injected spin polarized QP’s will be withdrawn by the second electrode, and thus spin accumulation will be zero. On the other hand, if the electrodes are in the AP-state, a finite spin accumulation will be induced. Hence, the suppression of superconductivity is not caused by the induced spin...
accumulation, but rather by the high density of QP’s in S, as shown in [99].

The superconducting nanowire can be driven normal by injecting a higher current density, as shown in Figure 8.1 (d). This allows us to reliably control the relative orientation of the magnetization of the F electrodes, using a high current injection (1-10 μA) and an externally applied magnetic field. This field is then switched off and measurements are performed with typical injection currents of 1-10 nA. Thus, the superconductivity is un-affected directly by the magnetic field or QP induced pair breaking, and we ensure that the effects discussed below originate from spin dependent transport.

8.2 Spin Signal and Spin Diffusion Length in a Superconductor

As discussed in Spin accumulation and relaxation in a superconductor, the spin signal is expected to increase at low temperature, for injection energies close to ∆. This is indeed observed, as shown in Figure 8.2, presenting the spin signal at a distance of 300 nm from the injection point as a function of bias current, for different temperatures. As the temperature is lowered just below $T_C \approx 1.5 \text{ K}$, $R_S$ increases three times, approximately to 1 Ω, but is independent of bias current. This is not shown in the plot, due to its small magnitude. Decreasing the temperature further, down to 300 mK, results in a dramatic enhancement of $R_S$ at low bias. For the lowest bias, $I_{inj} = 1 \text{ nA}$, $R_S$ reaches $\approx 400 \text{ Ω}$, which is more than three orders of magni-

![Figure 8.2: Spin signal at $x = 300$ nm as a function of bias current, for different temperatures. The lines are guides to the eye.](image-url)
CHAPTER 8. RESULTS IN THE SUPERCONDUCTING STATE

Figure 8.3: (a) Spin signal at 300 nm as a function of temperature, for different samples. (b) Normalized spin signal as a function of normalized temperature for sample 1, where $T_C \approx 1.6$ K and $R_S^{(N)} \approx 50$ mΩ. The line corresponds to the theoretical fit according to Eq. 4.24.

The measurements have been performed on several samples at low temperatures. At 22 mK and 1 nA of bias current, $R_S$ is of the order of kΩ, as shown in Figure 8.3 (a). For a more straightforward comparison, the normalized spin signal $R_S^{(S)}/R_S^{(N)}$ is plotted against the normalized temperature $T/T_C$, in Figure 8.3 (b). The line corresponds to a theoretical fit using Eq. 4.24. For sample 1, $R_S$ is enhanced by more than 4 orders of magnitude compared to the $R_S$ in the N-state. This is by far the largest $R_S$ measured in a metal/oxide nano-structure.

Recent theories by Takahashi and Maekawa [64, 102] and Morten et. al. [103, 104] predicted $R_S$ to diverge in the S-state. The former theory considers spin relaxation due to spin-orbit scattering, whereas the latter one also takes into account scattering from magnetic impurities. Following the approach by Morten et al., we found that the theoretical fit coincides well with the experimental data for temperatures down to $T \sim 0.2 T_C$. At that point, our measured $R_S$ starts to level off, which is believed to be due to a higher effective QP temperature than that given by the thermometer. Nevertheless, the giant enhancement of 4-5 orders of magnitude is a strong confirmation of the recent theoretical predictions on spin transport in superconductors [64,104]. The fitting parameter $\beta$ being 0.5 indicates that the dominant spin relaxation mechanism is due to magnetic impurities.

The QP’s are relatively decoupled from the phonon bath at low temperatures, which makes them difficult to cool down. An independent experimental evidence...
8.2. SPIN SIGNAL AND SPIN DIFFUSION LENGTH IN A SUPERCONDUCTOR

Figure 8.4: (a) I-V curve a Co/AlO$_2$O$_3$/Al junctions, measured at 22 mK. (b) Normalized differential conductance curve, $dI/dV$, together with a theoretical fit according to Eq. 4.15. The best fit was obtained for $T \approx 0.2 T_C$ and $\Delta \approx 200 \mu$eV.

for this temperature offset is obtained from measurements on the injection junction. Figure 8.4 (a) shows a typical I-V curve for the junction, measured at 22 mK. It demonstrates the feature of the superconducting gap, which is estimated to be $\Delta \approx 200 \mu$eV. By fitting Eq. 4.15 to the normalized differential conductance, shown in Figure 8.4 (b), we can extract the effective QP temperature. The best fit was obtained for $T \approx 0.2 T_C$. This is consistent with the saturation behavior of $R_S$, further supporting our interpretation.

The behavior of the spin diffusion length in superconductors, $\lambda_{sf}^{(S)}$, has been analyzed in recent theories [64,100,102–104]. It was predicted that $\lambda_{sf}^{(S)}$ increases with decreasing temperature [100], is the same as the spin diffusion length in the N-state, $\lambda_{sf}^{(N)}$ [64,102], or is significantly reduced for temperatures below $T_C$ [103,104]. Our experiments are consistent with the latter prediction by Morten et. al. and are illustrated in Figure 8.5 (a). The data clearly shows the decrease of $\lambda_{sf}$ with decreasing temperature, almost by a factor of ten at 20 mK compared to its value above $T_C$. Figure 8.5 (b), shows the normalized spin diffusion length, $\lambda_{sf}^{(S)}/\lambda_{sf}^{(N)}$, as a function of the normalized temperature $T/T_C$, together with a theoretical fit using Eq. 4.24. Our experimental data are well described by the theory, and the best fit is obtained for $\beta = 0.5$. This gives $1/\tau_m = 3/\tau_{so}$, which means that spin flip scattering by magnetic impurities is three times more likely than spin-flip scattering by spin-orbit interaction. With $\beta = 0.5$, the renormalization of the scattering rates described by $\alpha$ (Eq. 4.24) yields a diverging spin flip rate at $T \approx 0$, since the spins are injected close to the gap edge, i.e $E = \Delta$, where $\alpha = 0$. 

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Figure 8.5: (a) Spin diffusion length as a function of temperature, for 3 different samples. (b) Normalized spin diffusion length as a function of normalized temperature for sample 1 and 2, with $T_C \approx 1.6$ K and $\lambda_{sf}^{(N)} \approx 800$ and 600 nm, respectively. The line corresponds to a theoretical fit using Eq. 4.24.
Chapter 9

Results on the Spin Hall Effect in Superconductors

The strong reduction in $\lambda_{sf}$ in the S-state means that spin accumulation is short lived in S. In fact, the spin accumulation at 900 nm from the injector is negligible compared to the spin accumulation at 300 nm and 600 nm. This is illustrated in Figure 9.1, where $R_S$ at 600 nm and at 900 nm is plotted as a function of temperature. As expected, $R_S(600\text{nm})$ is decreasing with increasing temperature, whereas $R_S(900\text{nm})$ remains unchanged, lying within the noise level. Thus, QP's at 900 nm away from the injector and farther can be considered as completely un-polarized.

According to the theory described in Spin Hall effect (SHE), the spin Hall effect (SHE) signal, $\Delta R_{SH}$, is expected to diverge as the temperature approaches zero. Figure 9.2 shows the normalized SHE signal, $\Delta R_{SH}/\Delta R_{SH}(T_C)$, as a function of normalized temperature, $T/T_C$, using Eq. 5.5. The contribution from side jump

Figure 9.1: Spin signal, $R_S$, measured at 600 nm (full circles) and 900 nm (empty circles), with $H = 0$, as a function of temperature.
CHAPTER 9. RESULTS ON THE SPIN HALL EFFECT IN SUPERCONDUCTORS

Figure 9.2: Theoretical prediction for the temperature dependence of the normalized spin Hall effect signal in a superconductor, \( \frac{\Delta R_{SH}}{\Delta R_{SH}(T_C)} \), due to side jump (SJ) and skew scattering (SS). The signal is expected to diverge as \( T \) approaches zero [128].

(SJ) and skew scattering (SS) is taken into account individually. \( \Delta R_{SH}(T_C) \), corresponds to the SHE signal in the N-state, induced by a current driven through the Al wire.

Measurements of the temperature and bias dependence for \( \Delta R_{SH} \) are illustrated in Figure 9.3, where the lines are guides for the eye. As the temperature is decreased, \( \Delta R_{SH} \) increases significantly until it levels off at \( \approx 0.1 T_C \), which is attributed to the effect of the electromagnetic environment in the measurement system. Measurements and theoretical modeling of the differential conductance for the injection junction revealed the effective QP temperature to be \( \approx 0.3 T_C \) for this sample. This explains the saturation of \( \Delta R_{SH} \).

The DOS for the QP’s is enhanced by the opening of the gap, in which the highest density is obtained for injection energies close to \( \Delta \). From Figure 8.1 (a), we determine that a bias current of \( \sim 1 \) nA is optimum in our case for producing giant SHE signals. \( \Delta R_{SH} \) reaches values of the order of \( \sim 1k\Omega \) at \( \sim 1 \) nA of injection current. Increasing the injection current to 10 nA, results in a similar temperature dependence for \( \Delta R_{SH} \), but with a smaller magnitude. It also correlates with the decrease of \( R_S \) with temperature, shown in Figure 8.3, since superconductivity is suppressed by increasing the temperature. The SHE signal is more than 5 orders of magnitude higher than the SHE signal measured in the N-state in recent experiments [16,19]. It is obvious that the experimental data deviate from the theoretical prediction at the lowest temperatures. However, they are in qualitative agreement above \( \sim 0.28T_C \), confirming that a dramatic enhancement of \( \Delta R_{SH} \) takes place in
superconductors.

Another way of suppressing superconductivity is by injection of high energetic QP’s [99]. In combination with the DOS for the QP’s being smaller for these energies, $\Delta R_{SH}$ is expected to decrease. Figure 9.3 (b) shows $\Delta R_{SH}$ at 50 mK, as a function of bias current, which is proportional to the injection energy. Clearly, $\Delta R_{SH}$ decreases with increasing bias current. Observe that $\Delta R_{SH}$ lies within the noise level ($\sim 5 \Omega$ in this case) for currents higher than 100 nA, i.e the value sufficient to completely suppress superconductivity [99]. This correlates with the decrease of $R_S$ with bias current, shown in Figure 8.2, further supporting our interpretation. Thus, we have demonstrated the direct SHE in a mesoscopic superconductor.

Figure 9.3: Temperature dependence (a) and bias dependence (b) for the spin Hall effect (SHE) signal, $\Delta R_{SH}$. The lines are guides to the eye, and $T_C \approx 1.6$ K.
Chapter 10

Conclusions

This work investigates spin transport in mesoscopic conductors. A fundamental property of diffusive transport channels, serving as the foundation for theoretical modeling of the spin transport effects involved, is demonstrated on the experiment using a novel multi-terminal nano-device with a point like injection and symmetric spin sensitive detection about the injection point. It is shown that, within the experimental accuracy of better than 1%, spin propagation is governed by self-diffusion, independent of the profile of the underlying charge current. This should be significant for designing future lateral spin transport devices, which can have rather complex shapes of the spin transporting channels.

Particular emphasis herein is put on studying the spin relaxation mechanisms present in normal and superconducting mesoscopic conductors. Thus, a significant if not dominating contribution to spin flip scattering is expected for thin transport channels. For thicknesses comparable or thinner than the mean free path, scattering at the surfaces significantly reduces the Drude momentum relaxation time, which in turn should lead to a shorter spin relaxation time. This is certainly true for bulk impurity scattering, where the two relaxation times are directly correlated. We show that this correlation is not necessarily present in mesoscopic conductors and surface scattering. Depending on the band structure effects or a different nature of the impurities at the surface, surface spin flip can be substantially reduced. This result should be important for future nano-devices based on spin, where potentially even highly diffusive channels can be designed to have desirable spin transport properties using surface and interface optimization.

Mesoscopic superconductors are of high interest as spin transporting channels as they are expected to effectively amplify spin accumulation, theoretically by many orders of magnitude or even in a diverging fashion in the limit of zero temperature. The reason is the peculiarity of the electronic structure of a superconductor for injection energies near the gap energy. We provide the first demonstration of such an amplification, obtained in a direct electrical measurement.

Additionally, by observing and theoretically analyzing the decay of the spin
accumulation in a superconducting nanowire, we are able to differentiate the spin relaxation mechanisms present in the system. The differentiating point here is the different temperature dependence of the spin-orbit versus magnetic impurity scattering. This example of spin-flip spectroscopy across the normal-superconducting transition can be interesting for future characterization and analysis of materials used as channels in lateral spin devices.

A novel multi-electrode nano-device is used to provide the first demonstration of the Spin Hall Effect in a superconductor. Again the effect is demonstrated through a direct electrical measurement. The device has two branches, with designed to characterize in terms of the underlying spin transport, and the other branch used to detect the peculiar spin-orbit effect due to the long lived quasiparticles in the transport channel. Owing to the spin amplification property of the superconductor outlined above, the spin signals involved are record high, $\sim 1 \text{k}\Omega$. This result opens up many possibilities as two new mesoscopic transport effects and devices.

These five new or novel results address some of the key issues at the forefront of today’s spin-electronics research. The work presented should be technologically relevant through the basic knowledge generated.
Chapter 11

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Bibliography


BIBLIOGRAPHY


Chapter 12

Appended papers


5. N. Poli et. al., “Giant spin Hall effect in a mesoscopic superconductor.”, *manuscript.*

My contribution to the papers:

- In paper 1, I fabricated the samples and conducted the measurements. I analyzed the data and had the main responsibility in the manuscript preparation.

- In paper 2, I fabricated the samples and conducted the measurements. I contributed with the analysis of the data and took part in the manuscript preparation.

- In paper 3, I contributed with the sample fabrication and measurements.

- In paper 4, I fabricated the samples and conducted the measurements. I analyzed the data together with the co-authors, and had the main responsibility in the manuscript preparation.
• In paper 5, I fabricated the samples and conducted the measurements. I analyzed the data together with the co-authors, and had the main responsibility in the manuscript preparation.